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PROCEEDINGS OF THIRD INTERNATIONAL CONFERENCE ON FLUIDIZED-BED COMBUSTION

NATIONAL ENVIRONMENTAL RESEARCH CENTER

DECEMBER 1973

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16. ABSTRACT

The Third International Conference on Fluidized-Bed Combustion was held October 29-November 1, 1972, at Hueston Woods Lodge, RFD No. 1, College Corner, Ohio, under the sponsorship of the U. S. Environmental Protection Agency (EPA) The keynote address was, "The Clean Fuel Technology Gap-- Opportunities for New Fluidization Procedures." The themes of the six sessions are as follows: (1) Coal Combustion and Additive Regeneration; (2) Non-coal Fluidized-Bed Combustion Processors; (3) Gasification/Desulfurization; (4) Conceptual Designs and Economics; (5) Pilot Plant Design, Construction, and Operation; and (6) panel discussion summarizing the thoughts presented during the conference. All papers presented during the conference are included in these proceedings. Papers contains numerous figures and tables. All thirty papers together with the panel discussion and summary session are included in these proceedings.

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17. KEY WORDS AND DOCUMENT ANALYSIS

a. DESCRIPTORS		b. IDENTIFIERS/OPEN ENDED TERMS	c. COSATI Field/Group
Coal	Fluidized-bed processors	FBB (Fluidized-Bed Boilers)	
Gasification	Combustion chambers	Pressurized combustion	
Desulfurization	Manufactured gas	Clean fuel	
Economic factors	Electric power plant	New fluidization procedures	
Incinerators	Pilot plants	SGP (Shell Gasification Process)	
Limestone	Emissions		
Sulfurs	Air pollution		
Sorbents	Boilers		
Fuel oil	Regeneration (engineering)		10A 10B 13B
	Sulfation		

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PREFACE

The Third International Conference on Fluidized-Bed Combustion was held October 29-November 1, 1972, at Hueston Woods Lodge, RFD No. 1, College Corner, Ohio, under the sponsorship of the U.S. Environmental Protection Agency (EPA).

The Conference, under the general- and vice-chairmanship of Messrs. P.P. Turner and D.B. Henschel, respectively, was open to early arrivers Sunday, October 29.

The Conference proper, consisting of six sessions, got underway Monday morning with the official welcome extended by EPA's Robert P. Hangebrauck. Following Mr. Turner's introductory remarks, Dr. A.M. Squires delivered the keynote address, "The Clean Fuel Technology Gap — Opportunities for New Fluidization Procedures."

The six-part Session I, chaired by Mr. A.A. Jonke, followed the theme, "Coal Combustion and Additive Regeneration." Session II, held Monday evening, consisted of five presentations on the general topic, "Non-Coal Fluidized-Bed Combustion Processes;" Mr. Alvin Skopp was chairman.

Dr. Everett Gorin was chairman of Session III, titled "Gasification/Desulfurization," which opened Tuesday's activities. The session consisted of six presentations. "Conceptual Designs and Economics" was the theme of Session IV. It was chaired by Dr. D.H. Archer and consisted of seven papers.

Wednesday began with Session V which presented six papers on the topic, "Pilot Plant Design, Construction, and Operation." The Session was chaired by Mr. H.B. Locke. Session VI, the last of the Conference, was a Wednesday afternoon discussion, led by a panel of six, summarizing thoughts presented during the Conference and providing a final opportunity for comments from the floor. Chaired by Mr. Hangebrauck, the panel consisted of Dr. Archer, Professor D.E. Elliott, Mr. Henschel, Mr. H.B. Locke, and Dr. Squires.

All papers presented during the Conference are included in these proceedings.

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INTRODUCTORY REMARKS:

Mr. P.P. Turner, EPA

KEYNOTE ADDRESS:

Dr. A.M. Squires, City College of the City University of New York

1. THIRD INTERNATIONAL CONFERENCE ON FLUIDIZED-BED COMBUSTION

R. P. HANGEBRAUCK

Environmental Protection Agency
Control Systems Laboratory

I feel honored this morning to welcome on behalf of EPA all those attending the Third International Conference on Fluidized-Bed Combustion. A special welcome goes to those of you here from other countries, old friends and new alike.

Each time at the end of our Conference we have asked the attendees and ourselves whether it was worthwhile to assemble as we have here in the hills of Ohio, and each time I believe we have come up with a very positive yes. Aside from the up-to-date information exchange, it seems that a critical mass of expertise is achieved at this meeting causing significant reaction to occur during the meeting and long after.

The aim in our work on fluidized-bed combustion and in our Conference is to develop environmentally and economically sound systems for steam and power generation which will enable them to meet new source performance standards and ambient air quality standards for sulfur oxides, nitrogen oxides, and particulates, as provided by the Clean Air Act. Such systems must be compatible with these and other present and future environmental considerations, including water, land, heat, and noise pollution.

A variety of technologies for controlling pollution from stationary combustion sources will be forthcoming and should fit together like pieces of a complex puzzle to solve the applications problem in a cost and time optimized fashion. The technology we are working on here today is most directly applicable to the power industry, but because this technology will allow the use of lower cost dirty fuels and problem fuels, it will free clean fuels for smaller residential, commercial, and industrial fuel users constituting area-type combustion sources.

The projected application of technology and fuel resources will be such that, initially, part of the utility clean-fossil-fuel energy gap will be filled by increased use of low sulfur coal, physically cleaned coal, low sulfur and desulfurized oil, and most critically, flue gas cleaning systems which will act as a counter balance to prevent the demand pressure for clean and/or cleaned fuels from driving fuel prices too high. From now into the 1980's, the increased demand for electrical energy generated from fossil fuel, the shortage of naturally occurring clean fuels in the users locations, and improved economics for flue gas cleaning should cause a great expansion in the application of these systems. High-Btu gas and liquids from developing coal conversion systems will be limited to combustion sources considerably smaller than utilities, whose consumer classification is such that it justifies the inherent, much higher price. These processes should be commercially significant in the 1980's in filling the gas and petroleum gaps, but they will have their own set of environmental problems.

We realize that even though the needs for effluent cleaning will be satisfied by such measures, the ultimate extent of environmental control and lowest cost will not have been achieved. Considering the size and growth of the power industry and the multibillion dollar annual control costs involved, the public, government, and industry alike can no doubt see the potential payout for more effective, lower cost technology.

Fluid-bed combustion systems hold great promise for reducing or eliminating the excess costs created by the technology gap. The systems unfortunately are not here today, but are under development and nearing demonstration, as will be evidenced by the nature of our meeting here this week. We feel reasonably confident the technology will be available on a commercial basis as we move into the end of this decade. Aside from the built-in low-pollution nature of such systems for several pollutants, their cost effectiveness should cause a rapidly expanding share of the U.S. power-generating base to be filled by straight fluid-bed combustion systems and advanced power cycle gasification systems incorporating limestone/dolomite fluid-bed technology. To this end, EPA has invested considerable funds over the last five years, an investment which should be dwarfed, if successful, by the payout to the public and industry for more cost-effective use of fuels in generating power. I trust the progress we will hear reported this week will bear this out.

EPA program plans rely on an increased shouldering of the cost of development and demonstration by industry as the scale and cost of systems increases in the final stages of development. Considerable effort has been made in the EPA sponsored work to concentrate on the most promising approaches, and this has been caused in no little way by the small amount of funding available. However, it is hoped that each promising avenue will be explored at least somewhere in the world. It is also hoped that all information available can and will be used by other groups in a way that will prevent large expenditures of funds on scale up of systems of questionable environmental merit.

In any event, if we are to avoid getting into an expensive technology gap as new and more restrictive emission standards are set consistent with the health and welfare of this Nation and the world community, we will have to continue to look and move ahead with vigor on more productive and pollution-free processes.

Once again we welcome you to the Conference and encourage the fullest participation possible.

Thank you.

2. EPA-CSL PROGRAM TO CONTROL POLLUTION FROM STATIONARY SOURCES

P. P. TURNER

Environmental Protection Agency
Control Systems Laboratory

I would like to take this opportunity to repeat Mr. Hangebrauck and once again welcome each of you to the Third International Conference on Fluidized-Bed Combustion.

As was the case with the two preceding International Conferences, the purpose of this meeting is to bring together workers in the field of fluidized-bed combustion, and related areas, in an atmosphere conducive to cooperative information exchange. In addition, we have a number of organizations represented here which, although not currently directly involved in the research and development effort, will become involved in manufacturing or operating fluidized-bed boilers and their auxiliaries, in designing fluidized boiler plants, or in performing some other necessary function, when this promising new combustion technique is commercialized in the future. It is hoped that by getting all of you together, discussing your individual projects, and applying your individual expertise in this area, each of us can go home better informed of the overall international effort and with new ideas for direction of our individual efforts.

A large amount of work has been completed since the Second International Conference was held over two years ago. For our part, EPA has now spent a total of $7.6 million on work related to fluidized-bed combustion through the end of fiscal year 1972, of which about $5.2 million was committed since the last Conference here at Hueston Woods. Also during the past two years, design has been completed and construction begun on a 630-kW continuous fluidized-bed combustion pilot plant, capable of operating over the full range of conditions of interest, including pressure.

This Miniplant is designed to continuously regenerate the partially-spent SO_2 control sorbent generated in the fluidized combustor, and return the regenerated material to the combustor for re-use. Studies on the Miniplant will enable EPA to obtain answers for a number of important outstanding questions, and will provide the continuous combustion-sorbent regeneration data required to design the 20- to 30-MW pilot plant envisioned as the next stage of the development effort.

Also within the past two years conceptual designs have been completed for a 30-MW industrial coal-fired fluidized boiler, and for 300-MW and 600-MW utility-scale coal-fired fluidized boilers both at atmospheric pressure and at 10 atm pressure.

EPA's emphasis has turned toward fluiddized boilers operating at elevated furnace pressures, although atmospheric-pressure systems are also being evaluated. We are taking a close look at continuous regeneration of sorbent sulfated in the combustor, but operation with a once-through sorbent nonregenerative system has not been ruled out. During the past two years, a fair amount of evaluation has been conducted, and data have been generated, regarding pressurized operation and sorbent regeneration, in preparation for, and complementing, the forthcoming Miniplant program.

Development of the chemically active fluidized-bed (CAFB) atmospheric-pressure add-on residual oil gasification system, currently in the 750-kW stage, has advanced to the point where utility partners are being sought to provide a site for a 82-MW prototype installation.

Work is also continuing at an increased scale toward the development of a high-temperature fuel-gas desulfurization process to produce clean low-Btu fuel gas from caking bituminous coals.

All of this research and development effort sponsored by EPA will be described during the course of this meeting by the individual contractors involved. We also look forward, of course, to the discussions of work being conducted by organizations other than EPA in these and related areas.

Expressing the hope that the activities of this conference during the next three days will lead to the healthy exchange of information between all the participating members, and to numerous individual contacts, I declare this Third International Conference on Fluidized-bed Combustion open.

It is now my pleasure to introduce our keynote speaker who will address the conference on "The Clean Fuel Technology Gap: Opportunities for New Fluidization Procedures." He will review the problems which one sees ahead in the natural gas and petroleum markets; he will characterize the substitute technologies which will be needed; and he will review advantages of new fluidization procedures for treating coal and residual oil.

He is indeed well qualified to set the tone for this conference. Though his formal background is in chemistry, his interest in engineering was aroused during World War II through his association with Dr. Manson Benedict, whom he assisted in the process design of the Oak Ridge gaseous diffusion plant.

After the war he was Director of Process Development at Hydrocarbon Research, Inc. until 1959, when he resigned to become an independent consultant. He joined the faculty of the Department of Chemical Engineering of The City College of The City University of New York in September 1967, and was named Chairman of that Department in the fall of 1970.

He has published extensively on fluidization, oil and coal gasification, fuel desulfurization, gas cleaning, and power generation; he holds 15 U.S. patents in these fields. He has conducted research at The City College under EPA grants relating to the development of new systems for generating clean power from fossil fuels. His team at The City College began work last June on the first 18 months of a 5-year effort under a grant from the National Science Foundation to support "studies toward improved techniques for gasifying coal." The objective of these studies is to convert coal into pipeline gas and a light aromatic liquid fuel as well as low-Btu gas for power generation. Gentlemen, I present to this conference Dr. Arthur M. Squires.

Keynote Address

3. CLEAN FUEL TECHNOLOGY GAP: OPPORTUNITIES FOR NEW FLUIDIZATION PROCEDURES

A. M. SQUIRES

The City College of the City University of New York

Before we take up the specific problems of interest to this Conference, we may well first view these problems in the broader context of the markets for clean and convenient energy. To do so even briefly creates a sense of urgency and a demand for boldness. The World's appetite for clean fuels is sharply rising. That the World's resources of cheap clean fuels are finite is an inescapable fact, and economic consequences of this fact are beginning to be felt.

Consider Figures 1 and 2, which depict broadly the supply and demand situation for oil and natural gas in the United States between 1955 and 1985. The range of uncertainty in demand beyond the present is approximately the range among recent projections for 1985.

Figure 1 implies that America's economic growth is in jeopardy if we cannot import from one-half to two-thirds of our oil supplies in 1985. Imports to reach the upper curve amount to substantially the entire present output from the Middle East. Notice that oil from the North Slope has small relative effect. We would need to discover a North Slope each year between now and 1985 to reach the upper curve of Figure 1 from domestic supplies.

Gas cannot be imported from overseas as readily as oil, and Figure 2 implies a sharp flagging in the growth of the gas market. We are using a quantity of gas yearly that is greater than the average annual discoveries of gas made over the past 20 years and more than twice the discoveries of last year. No doubt discoveries can be increased by removing artificial restrictions on the price of gas at the wellhead, but no prompt effect on gas supplies could result. It takes several years to bring a new field to production, and in the meanwhile old fields decline. Nuclear stimulation is a doubtful proposition in light of concern over spread of radionuclides. All substitute natural gas from sources now in view will cost at least about \$1 per 10^6 Btu, and often more. This includes gas from the Far North. It should be remarked that not enough is known yet of North Slope gas reserves and production problems to project with assurance its supply to the United States market, yet some gas from the Arctic can probably be delivered by about 1980 with vigorous development, Canada willing.

Since much of the historic growth in demand for gas has been for fuel to fire boilers, a projection of the demand for clean boiler fuel would reveal a much greater gap between supply and demand than Figure 2 would suggest. The Nation wishes to eliminate emissions from boilers fired with coal and untreated residual oil. Satisfactory engineering solutions to the problem of ridding stack gases of sulfur dioxide are not yet in hand, and the problems of the large coal-fired power stations projected for the Southwest illustrate the dislocations of the forced shift from gas to coal in new plant construction.

An ironic illustration of the sharp change in the gas market is furnished by the news that

Coastal States Gas Producing Company will build a plant to manufacture synthetic natural gas from petroleum feedstocks in Texas.

It is hard to escape the impression that our energy markets will undergo price upheavals in the next decade. In view of this, it must seem astonishing to a layman that our fuel industries are so little prepared with substitute technologies. For example, to convert coal into clean gaseous or liquid fuels, they can absolutely rely, for immediate construction, only upon technology introduced nearly 40 years ago to fuel the German war machine. Moreover, nothing better will be ready to have much effect in the time span of Figures 1 and 2. Our research and development efforts, both private and governmental, have been far too inadequate. The layman must be further astonished to learn that private efforts are being reduced.

At least five major oil companies have shut down laboratories or layed off personnel engaged in synthetic fuels research. One architect-engineering firm that caters to the fuels industries has shut down an historically important laboratory, and other such firms appear to be decreasing their research budgets. Yet even a Manhattan Project on new fossil fuel technologies could not be expected to make a major dent in the clean fuel gap before 1985. It is highly improbable that synthetic liquid fuels made from coal could play a significant role before then. Most substitute natural gas will be made from petroleum feedstocks, which will themselves become short in supply. The extent to which we have been minding the store, in this particular respect, is ironically illustrated by the fact that we will license the SNG processes for petroleum feedstocks from Great Britain, Japan, and Germany at fees that will total more than $100 million.

It will be important to understand how the clean fuel technology gap has arisen. To what degree has research and development failure been due to governmental interference with "normal" economic processes? To what degree is it due to size and maturity of our energy industries and concomitant risks and high costs of research and development for these industries? Is it due to a general loss of appetite for risk-taking among the managers of our technology? Is it due to a general migration of creative talent, promotional and managerial as well as technological, into glamorous activities such as the space effort? Are there other factors?

Understanding these questions will be important, not only so that our domestic arrangements for fuels research and development may be overhauled, but also so that we may better prepare ourselves for an even more serious clean fuel technology gap which lies in the not distant future.

Figure 3 gives a gloomy but plausible scenario for the future course of the world petroleum market. If the attitude of the World toward oil parallels that of the United States toward natural gas, something like the scenario of Figure 3 will inevitably unfold. Substitutes for oil will be developed too late; production of oil will reach the limits of the World's capability to yield oil before sufficient experience with substitute technologies has been acquired; growth of technologies dependent upon oil will be choked off; and economic disarray as well as insufficient experience will prevent rapid growth of substitutes.

The historic excess in our capacity to produce gas, seen in Figure 2, was important to the petrochemical industry in the United States. Disappearance of the excess capacity, along with unwise import regulations for light hydrocarbon feedstocks, is creating serious difficulties for this industry, which illustrate in miniature the dampening effect that the disappearance of excess oil production capacity will have upon invention and development of new technologies for better use of oil. Existing equipment and existing technologies will preempt supplies, and opportunities to divert oil from uses of lower to higher value will be missed.

Electricity from whatever source (nuclear fission or fusion, solar, or geothermal) cannot be readily substituted for clean, portable fuels. Estimates of the U.S. fuel mix in the year 2000 postulate only about 25 percent nuclear at best. For the distant future, electricity could electrolyze water to yield hydrogen. This could be used directly as a fuel, stored either as liquid or at high pressure or reversibly adsorbed upon a solid. Alternatively, hydrogen and carbon dioxide could be converted to hydrocarbons by Fischer-Tropsch synthesis, or hydrogen and nitrogen could be converted to ammonia. By what steps and over what kind of time span might such a synthetic fuel economy be introduced? In an early transitional stage, electrolytic hydrogen might add to the heating value of carbon-rich fuels of natural origin, converting them to lighter materials. Even earlier, if natural gas is still available in regions like Venezuela and the Middle East, hydrogen might be made from the gas and added to carbon-rich fuels.

To what extent can the world rely upon synthetics based upon coal? It should be noted that the Northern Hemisphere is much richer in coal than the southern. Even in the Northern Hemisphere, coal deposits are concentrated in a few favored countries (notably the United States and Russia, especially the latter). If SNG from Texas is a surprise, what about exports of synthetic liquid hydrocarbons from Wyoming? Our coal resources seem vast, but would they if measured against the projected world appetite for oil in the 21st Century?

What other substitute technologies based upon electricity can replace liquid fuels? Can nuclear energy (or solar or geothermal) be increased beyond present projections before the year 2000? On the other hand, what happens if our second try for a liquid-metal-fast-breeder-reactor is a flop, as was the first?

Can solar-pond algae plausibly contribute to the gap? Hydrocarbons based upon human and animal wastes? Cellulose? What about the "substitute" of making do with less? If there must be a belt-tightening anyway, it would be better earlier than later.

Can a combination of these or other developments, carried out in a timely manner, produce the more attractive scenario of Figure 4?

Figures 3 and 4 carry no scale for time. Hubbert has drawn his celebrated "dimple" for two estimates of "recoverable" world oil reserves (meaning recoverable at costs not far advanced from current costs): 1350 and 2100 x 10^9 barrels. At the lower figure, production begins to depart from a substantially exponential rate of growth at around 1980 and reaches a peak at about 66 x 10^6 bbl/day in about 1990. At Hubbert's higher figure, he projects a "dimple" with substantially exponential growth until about 1990 and a peak production at about 102 x 10^6 bbl/day shortly after the year 2000. Current production is nearing 50 x 10^6 bbl/day, and appears to be ahead of Hubbert's projections.

At Hubbert's lower estimate of reserves, the crisis in Figure 3 could come as early as about 1985. At the higher estimate, about 2000. This is assuming at least modest cooperation from both oil-producing nations and authorities responsible for leasing off-shore drilling sites.

The key to the scenario of Figure 4 is timely research and development and timely construction of full-scale installations in which to practice the new technologies. It is already too late to prepare for the world clean fuel crisis if it should appear as early as 1985. There is just barely time, perhaps, to get ready for problems that might reach critical proportions in 2000. Thirty years is not too long to develop and test a new technology and to learn it sufficiently well that it may be expanded as rapidly as the scenario of Figure 4 will require.

Energy decisions made in the United States in the next few years can be crucial in the choice of scenarios. That of Figure 4 may well imply the lower curves of projected fuel demand in Figures 1 and 2 as well as a

Manhattan Project to acquire good technologies for converting coal to gas and oil. Simultaneously, we must not neglect vigorous attack on the substitute technologies whose commercialization must begin in the 1980's if they are to be ready to forestall a world energy crisis.

OPPORTUNITIES FOR AGGLOMERATING AND FAST FLUIDIZED BEDS

Our first chapter has raised more questions than it has provided answers.

My second chapter, turning to the interests of this Conference must inevitably be a more particular response to the urgent call for new fossil fuel technologies.

My colleagues at The City College and I believe that the time is coming soon when economics will turn against the practice of burning chemically-bound hydrogen for large-scale production of electricity. Instead, the bound hydrogen in coal or residual oil will be viewed as too valuable to burn and send as water vapor up a stack. The hydrogen can become a part of some clean, convenient fuel having an economic value higher than coal's or residual oil's. The hydrogen-rich fuel would be "creamed off" the coal or residual oil leaving a carbon residue that would be burned to generate heat or electricity.

The idea of creaming off valuable products from coal or oil is of course not original with us. The byproduct coke oven is 100 years old; many attempts to displace it with improved coking procedures have been recorded; and the oil industry has made steady advances in technologies for reducing the yield of resid and increasing yields of lighter products. Most attendees at this Conference will be familiar with Consolidation Coal Company's efforts, as well as the recent achievements of the FMC Corporation.

Any good idea, however, can stand constant review in light of the appropriate technological context. We have tried to review the foregoing idea in light of the ongoing development to supply the military with better gas turbines, funded at about $300 million per year. Progress in engines for civil aircraft and stationary power has historically followed military achievements after a lag of only a few years. The 747 flies today with a temperature of 1300°C at the inlet to the turbines. Stationary machines larger than 100 MW are promised for this temperature before 1980.

Existence of such machines will allow design of electric power installations of sharply advanced efficiency and reduced capital cost. Gas turbines (inherently cheap in cost) will supply about 50 to 60 percent of the power, and a steam system (using modest steam conditions and a cheap boiler) will scavenge heat from gas turbine exhaust.

The prospect of these designs creates an imperative to develop better techniques for gasifying coal and residual oil to provide a cheap "power gas" to run the gas turbines—i.e., a low-Btu gas made using air and a little steam as the gasification medium. (Power gas was Ludwig Mond's term for producer gas, which he used to generate electricity in gas engines. He founded The Power Gas Company.)

The City College team believes that power gas technologies will inevitably evolve to allow some cream-skimming that pulls out fuel products of value greater than coal or residual oil. Nevertheless, power gas technologies will probably arise in the first instance for treatment of raw fuels. It is doubtful that at first any more than very slight consideration can be given to the potential of these technologies for profitable evolution. Let us first consider power gas technologies for raw fuels, and second, how they might evolve.

PRODUCTION OF POWER GAS

Among gas-to-solid contacting procedures, fluidization will be the strongest candidate for employment in better technologies for gasifying coal or residual oil. To a large degree, this is so simply because of the scale of the power stations to be built in the 1980's and beyond. Sites for over 4000 MW are already in service. Many more will be built. Typical sites will process coal, for example, on the scale of scores of thousands of tons per day; oil, at hundreds of thousands of barrels per day. Only fluidization procedures can readily provide equipment of capacities that will avoid the necessity of processing coal or oil in an unattractively large number of vessels operating in parallel.

Although the science of the gravitating bed is far ahead of our knowledge of fluidization, the former art is difficult to build in large capacities. The blast furnace, after more than a century of development, gasifies up to about 4000 tons of coke per day, but coke of course is a processed and closely-sized solid. More than 20 Lurgi gravitating-bed pressure gasifiers of the current design would be needed for 1000 MW; scale-up of this approach may prove difficult and uncertain. Although the gravitating-bed had significant yield advantages, it did not win in competition with the fluidized-bed catalytic cracker because it could not easily reach capacities appropriate to the scale of oil processing after 1960.

Gasification of either coal or oil in absence of a bed of solid appears plagued with a carbon loss problem which may inevitably require extra equipment for extinction of carbon. Either Texaco or Shell "partial oxidation" of residual oil must provide for carbon recovery and recycle to achieve complete carbon utilization. Nothing was published from Texaco's large experiment with a slagging, dilute-phase gasifier at Morgantown in 1957, but experience at Bell and elsewhere suggests that carbon utilization in such a gasifier may be poor, especially for a coal with a refractory ash.

I have not seen any advantage in gasification of coal in a pool of iron, a procedure of doubtful integrity at elevated pressure and doubtful operability at atmospheric pressure.

The City College view is that the strongest candidate for gasifying coal to obtain power gas will combine the ash-agglomerating fluidized bed about which we will hear from Godel and the circulating fluidized bed that Schmidt will describe. The combination would operate at about 2000°F and about 10 ft/sec velocity. A single vessel could easily handle coal for 1000 MW; at 20 atmospheres, the diameter would be less than 20 feet. The role of the ash-agglomerating bed would be to gasify large particles of coal, up to about 3/4-inch as well as to agglomerate and separate ash matter from the carbon-rich bed. At The City College we have re-dubbed Lurgi's highly expanded circulating fluidized bed the "fast fluidized bed;" we have a two-dimensional fast bed of plexiglas in operation that is exciting to watch. The fast bed in the gasifier combination would gasify fines and would provide a zone of high velocity and intense circulation for introduction of a caking coal. It is reasonable to hope that a caking coal could be successfully introduced into the fast bed, fine particles joining the bed and a large particle being coked sufficiently on its surface to render it harmless before it reaches the ash-agglomerating bed below.

Data by Dent, which I have discussed in "Role of Solid Mixing in Fluidized-Bed Reaction Kinetics" to appear in an AIChE Symposium Series volume, strongly suggest that the kinetic performance of the proposed combination will be excellent. Because of the high temperature and good kinetic performance, flow of steam will be small relative to air. Table 1 compares Lurgi gravitating-bed power gas with gas from the proposed gasifier calculated with the assumption that steam-carbon equilibrium is substantially achieved therein, an assumption suggested by Dent's experience. Table 2 compares electricity-generating efficiencies of combined-cycle installations (with power equipment according to United Aircraft's "second generation" design) using the Lurgi gasifier with wet gas cleaning, or the ash-agglomerating-fast-bed gasifier with wet cleaning, or the latter gasifier with gas cleaning by the high temperature procedures that we have under study at The City College.

Table 1. COMPARISON OF CRUDE POWER GAS FROM THE LURGI GASIFIER AND A CANDIDATE FOR DEVELOPMENT

Composition, % by vol	Lurgi gravitating-bed gasifier	Gasifier combining ash-agglomerating and fast fluidized beds
Methane	4.4	0.5
Carbon monoxide	10.7	31.8
Hydrogen	15.7	15.6
Carbon dioxide	10.7	0.5
Water vapor	27.8	0.5
Nitrogen	30.2	50.4
Hydrogen sulfide	0.5	0.7
	100.0	100.0
Heating value, Btu/ft ^{3}	129	157

The high loss of latent heat in the stack from a combined-cycle plant that depends upon the Lurgi is inherent for this gasifier. The loss arises from two sources: air supplied to the gasification bed is necessarily accompanied by a high flow of steam in order to limit the temperature and to keep the ash free-flowing; and gas leaving the bed must be quickly reduced in temperature by a water quench in order to prevent formation of heavy tars that would lead to deposits of coke.

A fluidized-bed gasifier operating at 2000°F will not make tars or tar-forming species, and its power gas need not be quenched.

One cannot be so confident in respect to a gasifier working at 1700°F. The Winkler did not make tars, but secondary oxygen was introduced into the Winkler above the fluidized bed in order to raise the temperature (to reduce methane yield? to eliminate tar-formers?). Late-model Winklers were pear-shaped and had enormous freeboard regions. For operation at 1700°F, a carbon-burnup step must be provided, as Pell will describe, and there will probably be a price also in capacity and in loss of latent heat to the stack, as well as a possible problem with tar-formers. Nevertheless, until development of a gasifier exploiting Godel's ash-agglomeration principle is far enough along, an approach at lower temperature is well worth carrying as an alternate.

Experience at Hydrocarbon Research, Inc.and elsewhere suggests that defluidization phenomena might plague an attempt to develop a gasifier to operate between about 1800 and 2000°F, unless perhaps the development were to adopt a fluidization velocity so high as to approach the fast-fluidized state. The limits would of course vary with properties of coal ash, and may extend below 1800°F for some important coals.

I have not yet seen advantage for an approach using an initial step carbonizing coal at about 1700°F followed by gasification at 2000°F, the flows of solid and gas between the two steps being countercurrent. During this Conference we will perhaps hear evidence supporting an advantage for this approach. However, there would appear to be a substantial risk that tar-formers will appear in gas from a carbonization step at 1700°F, requiring a rapid quench of the gas to prevent coke laydown in transfer lines. To avoid unnecessary degradation of heat, a quench is of course best avoided except in a case where a useful quantity of liquid-fuel byproduct can be recovered.

I have also not seen advantage for a separate ash-agglomerating zone such as Je'quier provided. This was a zone lean in carbon and was both more dilute and hotter than Je'quier's primary fluid bed. It afforded complete combustion of air furnished thereto, and it served to densify ash agglomerates and to reduce their carbon content. If reduction of carbon content in ash agglomerates should become a problem, Godel has provided two simpler approaches. His grate emerges from the bed, and agglomerates thereon are exposed to air in absence of coke particles external to the agglomerates. Godel has also demonstrated burnup in a gravitating-bed of agglomerates from an anthracite of high ash

Table 2. ILLUSTRATIVE ENERGY BALANCES FOR COMPLETE POWER-GENERATION FLOW SHEETS

Category of energy loss, %	Conventional steam power equipment without recovery of sulfur	Combined-cycle power equipment with recovery of sulfur[a]		
		Lurgi gasifier, gas cleaning at low temperature	Gasifier combining ash-agglomerating and fast fluidized beds	
			Gas cleaning at low temperature	Gas cleaning at high temperature
Electricity sent out	39.5	45.0	49.1	50.5
Heating value of sulfur	--		1.1	1.3
Loss of sensible heat in stack gases	5.0	4.6	4.5	4.7
Loss of latent heat (water vapor)	3.8	14.1	5.6	4.5
Loss of heat at steam condenser and elsewhere	47.7	28.4	35.7	35.0
Loss of unburned fuel and heat leakage	2.0	4.9	2.0	2.0
Mechanical losses and power for auxiliaries	2.0	2.0	2.0	2.0
	100.0	100.0	100.0	100.0
Efficiency, allowing credit for heating value of sulfur	39.5	45.5	49.7	51.2

[a] United Aircraft "second generation" design parameters.

content, the bed being supplied with air at the bottom. If carbon burnup resists these solutions, a separate burnup step exploiting Battelle's carbon-lean ash-agglomerating bed for complete combustion would be preferable to Je'quier's arrangement.

In light of work at Esso Abingdon by Moss and his colleagues, gasification in a fluidized bed containing lime emerges as a strong candidate for production of power gas from oil. Presence of tar-formers in the Abingdon gas gives cause for concern, however. Periodic burnout of coke deposits would not be an attractive procedure at high pressure. In a commercial embodiment of the Abingdon ideas, could the cyclone on the gasification bed be dispensed with? My thought would be to introduce secondary air directly above the gasification vessel at the elevation where the fuel gas enters the larger cross-section of the boiler being served. For production of power gas at high pressure, a comparable idea would be to introduce secondary air above the fluidized bed, raising the temperature of the power gas (eliminating tar-formers? as in the Winkler?). If this does not work, we might be forced to supply much more air than Abingdon uses to a gasification bed working at high pressure, thereby producing a leaner gas containing less hydrocarbon species. Heat-removal surface would need to be provided.

EVOLUTION TO THE FUELPLEX

W.C. Schroeder published data in his U.S. Patent 3,030,297 (April 17, 1962) that seem to The City College team to provide a strong lead in respect to a candidate for the first fuel-treating step in a "Coalplex" producing substitute natural gas, liquid fuel, and electricity. Solihull and Bruceton long ago taught that the treatment of raw coal with hydrogen at elevated pressure and at temperatures between about 1500 and 1800°F can result in attractive yields of methane at high concentration. In Solihull and Bruceton experiments, the residence time of vapor product generally ran into the minutes. Schroeder's contribution has been to call attention to the attractive yields of benzene, toluene, xylene, and substantially the same yield of methane, accompanied by very little heavy tar if the vapor product residence time is kept short, preferably around 5 seconds. At the long residence time of the earlier experiments, aromatic products polymerized, condensed as heavy tar upon the coke present in the reaction zone, cracked to form additional coke, and vanished. By arranging for a rapid quench of vapor species to about 700°F, Schroeder preserved tar-forming aromatic species and thereby obtained an attractive yield of a light aromatic liquid.

The City College team sees Schroeder's chemistry as opening up the possibility for a Coalplex yielding roughly 25 percent of the coal's heating value in form of methane and perhaps 15 to 20 percent as BTX, with the remainder being converted to electricity at an efficiency beyond 40 percent. The heat degradation that results from Schroeder's quench is tolerable, amply rewarded by the yield of liquid product that the quench preserves.

For Western coals, the fast fluidized bed is a candidate device for conducting Schroeder's chemistry. The fine coke product could be circulated from the fast bed to a heating step and returned to sustain the slightly endothermic Schroeder reactions.

For Schroeder's chemistry with Eastern coals, the coke-agglomerating fluidized bed that I discussed here during our Second International Conference is a candidate device, perhaps with a superposed fast fluidized bed of a fine solid that is circulated to provide heat.

For either coal, the coke product could be gasified in the aforementioned combination of Godel's ash-agglomerating fluidized bed and a superposed fast bed to deal with fines.

The coke product could also be burned up in a fluidized bed boiler. Although The City College team regards production of power gas to be the main chance, nevertheless we have a healthy respect for the difficulties of gasifier development, and the boiler is welcome competition. Thanks to Godel's 35 Ignifluid "gasifiers" (to call them "boilers" in this context is misleading), the race at the moment appears about even.

A counterpart of Schroeder's procedure as applied to residual oil should yield even higher quantities of methane and BTX and less coke. Coke from oil treatment is practically certain to contain negligible sulfur. In form of beads roughly 1/12 to 1/2-inch in size, the new petroleum coke, after calcination, should find lively markets for electrode manufacture and metallurgy.

Whither Fossil Fuel Development?

Our first chapter set down a gloomy picture of fossil fuel research and development. Past efforts, both private and governmental, have not been responsive to urgent needs, and the former are contracting.

Our second chapter, although it focussed upon The City College's view of the future, nicely illustrates the necessity for sharply expanded research budgets.

In perhaps no engineering procedure other than fluidization is art so far ahead of science. Neither ash-agglomerating, coke-agglomerating, fast fluidized beds, nor other competing procedures for processing coal will be developed through computer modelling or "optimization" studies. Godel discovered his ash-agglomeration phenomenon in 1954 during hands-on hardware development of processes for making activated carbon from coal. Although he had no experience with boilers, Godel promptly recognized a new capability to gasify and render burnable anthracite slacks from the mine at La Mure, near his boyhood home at Vif, Isere. With help from Babcock-Atlantique, he had a small boiler operating inside a year. Although the fast fluidized bed arose from Lothar Reh's work on fluidization in cones for his Dr.-Ing. Degree, Lurgi had to carry the development through frustrating difficulties as Reh's team learned how to achieve the fast-fluidized state in large-scale, practicable equipment. The origins of agglomerating beds that make dense beads of agglomerated material are obscure to me. Chance observation by Dorr-Oliver during development of a fluidized roaster for a "sticky" Katanga ore may have been important. Dorr-Oliver had a commercial process for calcining calcium carbonate slimes before 1957, and a number of pilot operations were underway at Dorr-Oliver, Fuller, and Battelle before 1960. Yet in 1972 we have hardly begun to acquire a scientific knowledge of these beds.

A number of factors have joined to create an illusion, shared by public and political leaders, research managers, and alas far too many engineers, that a new development requires only laboratory results and design of appropriate equipment based simply upon scientific analysis. One factor has been the dominance of the physics establishment in selection of recent national R&D goals. A second factor has been emphasis among educators upon engineering science to the detriment of an appreciation for hands-on development of new engineering hardware. The latter has somehow not been respectable by comparison with fancy mathematical analysis; it also gets a fellow dirty. A third factor, of special importance to the chemical engineering profession, has been the success of the approach exploited so brilliantly by Scientific Design Co., [illegible]. immediately after World War II; viz., bench development of fixed-bed catalytic processes followed directly by commercial-scale equipment. A literature has even grown up on the theme, "the pilot plant is obsolete." This situation may exist for fixed-bed processes in which there is no risk of minor unwanted products undetectable at the bench yet troublesome in the field, but this special case should not be elevated to a philosophy. Permit me a bit of personal history. I got an altogether false idea of engineering from my first job working on the design and startup of the first gaseous diffusion plant at Oak Ridge: this plant was built from bench data, was "scientifically designable," and was a justly celebrated success. I learned the other side of engineering through my participation in the classic flop of fluidized-bed process development. The hydrocarbon synthesis reactors failed at Brownsville because the development lacked a timely pilot unit of adequate size. If Dubie Eastman's 12-in. pilot plant at Montebello had operated 3 years sooner, we would have known. Our research budget had been too small.

Budgets unresponsive to our clean fuel needs have led to a preference for "safer" experiments relying more heavily upon earlier experience and representing a "simpler" approach. We have learned that the "simple" is not always so: witness the misfortunes of the dry-limestone-injection approach and the troubles of limestone scrubbing. In the meantime, for lack of making ourselves ready, unanticipated opportunities arise that cannot be seized. Studebaker-Worthington's Turbodyne and Southern California Edison are showing how old steam turbines can be converted to a combined cycle by scrapping old boilers and adding gas turbines followed by new waste heat boilers. General Electric and Westinghouse report brisk sales of combined-cycle equipment. A market for power gas is developing right under our eyes, and only Lurgi is ready for coal and, except Texaco-Shell, partial oxidation for oil.

In relation to the possibilities and the urgency of our needs, progress in areas to be covered during this Conference must seem disappointing to any visitor at Alexandria 5 years ago who admired the remarkable hands-on hardware development already accomplished by Pope, Evans & Robbins by that date; or, to a visitor to Leatherhead more than 3 years ago who saw BCURA's feat for burning coal at elevated pressure at rates approaching 1000 pounds per hour. Fluidized-bed boiler development in the United States needed Esso's "miniplant" 3 years earlier and 10 times bigger, but our hosts at this Conference simply have not had available to them the requisite funds. Nearly 3 years ago at the Christmas AAAS meeting in Boston (and on many occasions subsequently), I pointed out the potential value of Godel's ash-agglomeration phenomenon in combination with the fast bed in a maker of power gas, and I said it was a shame that we had no commercial experience here with the phenomenon. It is still a shame. Our fossil fuel industries have not had the simple curiosity to buy a small Ignifluid boiler, a commercial proposition on which nothing would be lost, to gain firsthand experience from its operation. Our hosts here have not had available funds to make good this omission.

Let me close by quoting from F.C. Dent's Melchett Lecture of 1965. This great development engineer, now enjoying life on his yacht out of Malta, can reflect with pleasure on the scores of millions in royalties that the SNG processes he developed at Solihull will bring during the next few years to the British economy. In terms of the quotation to follow, our own efforts to develop SNG processes for coal may be said to be just beginning.

> It is significant. . .that we usually had reason to regret any protracted period of exploratory laboratory investigation. Small-scale experiments have often been time-wasting even when large-scale conditions have been reproduced as faithfully as possible. . .Operation on a reasonable scale at an early stage is most desirable to throw difficulties into their proper perspective. Laboratory work was of most value after the problems had been recognized in this way.

All technologies addressed toward closing the clean fuel technology gap must include a major materials-processing step, handling solids, liquids, or gases on a scale almost without precedent in chemical engineering art. Serious effort does not begin until this step is addressed by hands-on hardware development on a practical scale. Budgets must be big. Programs must be bold.

APPENDIX

Energy Studies at The Department of Chemical Engineering of The City College of The City University of New York

Nine studies are in progress. All except III, IV, and IX are supported by Grant GI-34286 from the RANN Program (Research Applied to National Needs) of the National Science Foundation. Professors Michael Gluckman, Robert Graff, Robert Pfeffer, Reuel Shinnar, and Joseph Yerushalmi; Dr. Norman Holcombe; and Messrs. Samuel Dobner, Kun-Chieh Lee, Dennis Leppin, Jeffrey Silverstein, Eugene Yu, Stanley Dobkewitch, and Nurettin Cankurt are participating in the effort. In the past, Drs. Leon Paretsky, Melvyn Pell, and Lawrence Ruth, and Messrs. Richard Angiullo, Richard Barth, Ralph Levy, Basil Lewris, Michael Somer, and Lauris Sterns made substantial contributions. Messrs. John Bodnaruk, George DiIorio, Michael Askenazy, and John Spencer have helped with experimental arrangements.

Research on Power Gas:

I. Study of the Godel Ash-Agglomeration Phenomenon.

II. Study of Kinetics of Carbon Gasification in a Fluidized Bed. Our objective is to test F.J. Dent's hypothesis that the superior kinetics afforded by a fluidized bed for the steam-carbon reaction are a consequence of solid mixing in the bed, bringing about repeated exposure of carbon to fresh gasification medium.

III. Study of Kinetics of Removal of Sulfur Compounds from Power Gas by Action of Calcined Dolomite.

IV. Study of Removal of Fine Dust from Power Gas by a Panel Bed Filter. The filter can be built to clean gas at temperatures approaching 1800°F. We have achieved cleaning efficiencies beyond 99.99 percent for power station fly ash at a normal stack dust loading in small-scale tests at atmospheric temperature.

Research on the Coalplex:

V. Study of Reaction of Coal with Hydrogen at High Temperature and Pressure and Short Residence Time of Vapor Products. We believe this reaction, disclosed by W.C. Schroeder, can be the first coal-treating step in a Coalplex shipping substitute natural gas, BTX, and electricity at costs below the combined cost of making each product separately from coal.

VI. Study of Coke-Agglomerating Fluidized Bed, a candidate device for conducting Schroeder's chemistry (as in V above) on Illinois and other Eastern coals.

VII. Study of the Fast Fluidized Bed, a candidate device for conducting Schroeder's chemistry (as in V above) on Western coals; also, for gasifying fine particles of carbon blown from an ash-agglomerating fluidized bed.

VIII. Flowsheet Studies for the Coalplex.

Research on the Oilplex:

IX. Flowsheet Studies for an Oilplex in which oil is first treated by reaction with hydrogen at high temperature and pressure and short residence time of vapor products.

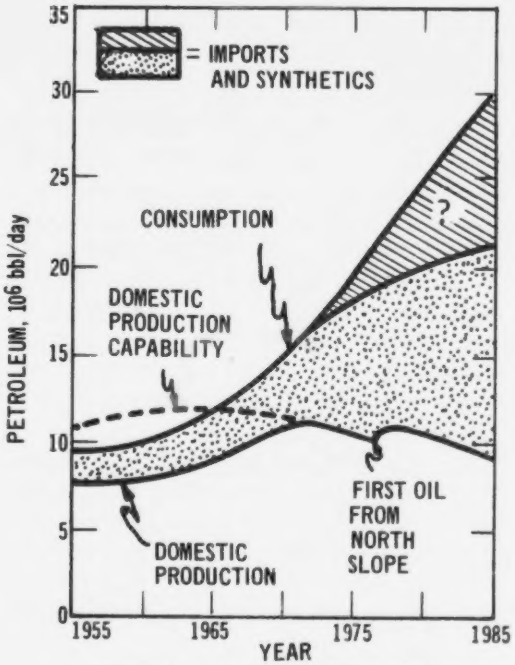

Figure 1. United States petroleum supply and demand.

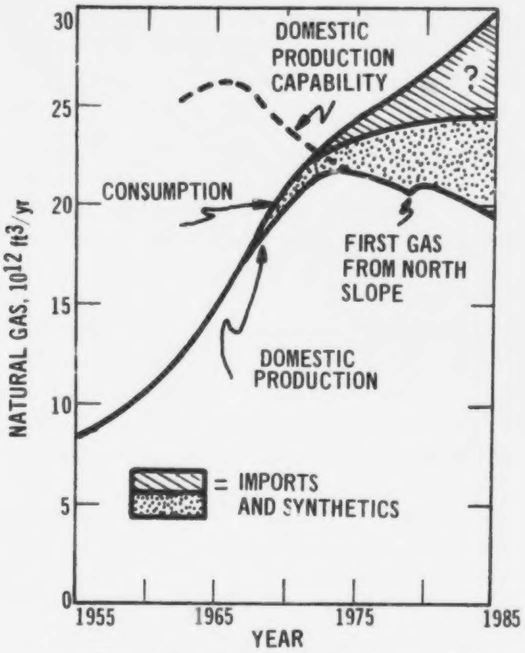

Figure 2. United States natural gas supply and demand.

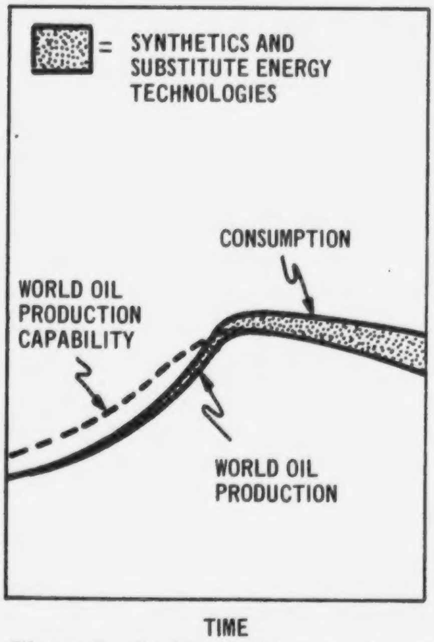

Figure 3. World petroleum supply and demand: a gloomy scenario.

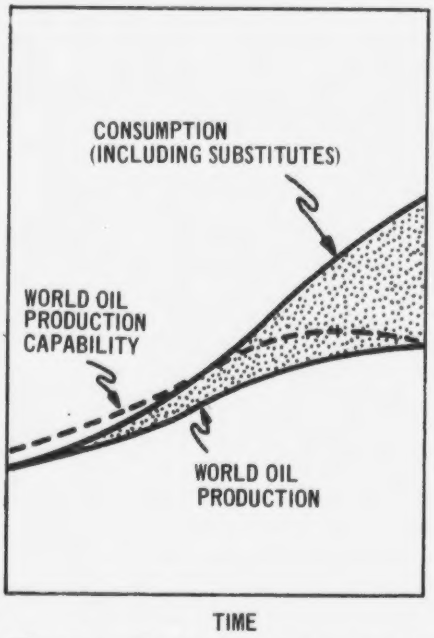

Figure 4. World petroleum supply and demand: another scenario.

SESSION I:

Coal Combustion and Additive Regeneration

SESSION CHAIRMAN:

Mr. A.A. Jonke, Argonne National Laboratory

1. BENCH-SCALE DEVELOPMENT OF COMBUSTION AND ADDITIVE REGENERATION IN FLUIDIZED BEDS

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ABSTRACT

This paper discusses information obtained since the last Hueston Woods Conference on the combustion of coal and oil with an excess of air and the combustion of coal in a deficiency of air. The paper is also concerned with the thermodynamics of several proposed regeneration processes and the regeneration of sulfur-containing additive by the two most promising processes — a one-step reductive decomposition of $CaSO_4$ and a two-step (reduction-CO_2/H_2O) procedure.

INTRODUCTION

Research on the combustion of fossil fuels (particularly coal) in a fluidized bed of solids is currently under investigation in the United States and other countries. Most of the U.S. effort is supported by the U.S. Environmental Protection Agency (EPA), Office of Research and Monitoring.

In applications of fluidized-bed combustion, fuel is burned in a fluidized bed of solids in which boiler tubes are immersed to take advantage of the high heat-transfer characteristics of fluidized beds. Additive, either crushed limestone or crushed dolomite, can be continuously fed to a fluidized-bed combustor to react with SO_2 released during combustion and provide a means of in situ control of the emissions of SO_2.

Two different combustion modes are possible, one with complete and the other with partial combustion of the fuel in the fluidized bed. In the complete-combustion mode (also called one-stage or oxygen-excess combustion), oxygen in excess of the stoichiometric amount required to burn the fuel to CO_2 and H_2O is added to the fluidized bed. In the second mode (called two-stage or oxygen-deficient combustion), a stoichiometric deficiency of air is added to the fluidized bed, and the resulting H_2, CO, and hydrocarbons are combusted to CO_2 and H_2O by providing additional oxygen (air) either in the region above the bed or in a separate combustor.

At Argonne National Laboratory (ANL), data have been obtained on combustion of coal and oil with an excess of oxygen and on coal with a deficiency of oxygen. All experiments have been made at atmospheric pressure. The objectives of these experiments have been as follows:

1. To determine how sulfur retention is affected by independent fluidized bed operating variables such as bed temperature, gas velocity, oxygen concentration, bed height, calcium to sulfur ratio, type of additive and coal, and additive and coal particle size.

2. To determine the level of NO in the flue gas at different operating conditions.
3. To obtain information on combustion efficiency, combustion products, limestone utilization, extent of calcination, decrepitation rates.
4. To obtain information on the mechanism of the lime sulfation reaction.

Efficient removal of SO_2 from the gas phase requires moderately large quantities of limestone (compared to the quantity of coal ash). When coal is burned, it will be desirable to regenerate the partially sulfated lime. Thermodynamic calculations and experimental data are presented on the two most promising reactions; i.e., high temperature (~2100°F) reductive decomposition of $CaSO_4$ and a two-step process, low temperature (~1600°F) reduction of $CaSO_4$ followed by reaction of the CaS with CO_2/H_2O.

BENCH-SCALE ATMOSPHERIC COMBUSTION EXPERIMENTS

Materials, Bench-Scale Equipment, and Procedure

Figure 1 is a schematic diagram of the bench-scale fluidized-bed combustor system. The combustor is a 6-in. diameter stainless steel vessel. The fluidizing air* enters the combustor through a bubble-cap air distributor mounted on the bottom flange. Feeding and metering of coal and additive is done by variable-drive volumetric screw feeders mounted on scales. The solids are fed pneumatically (entrained in a transport air stream) into the fluidized bed at a point just above the gas distributor.

Solids are removed from the off-gas by two high-efficiency cyclone separators in series and a glass fiber final filter. Downstream from the cyclones, approximately 20 percent of the total flue gas is diverted to a gas-analysis system and its water content is reduced to 3000 ppm (by condensation and refrigeration) to prevent moisture interfering with gas analysis. Continuous analyses of the dried gas for NO, SO_2, CO, CH_4, and O_2 are conducted with infrared analyzers and a paramagnetic oxygen analyzer. Gas chromatography provides intermittent analyses for CO_2. Periodically during a run, the bed and the overhead solids are sampled to permit chemical analysis and to obtain material balances. All instrument signals, pneumatic and electrical, are routed to a data logger which produces a paper tape record for further data processing and a typed output of the signal values.

In a startup, the fluidized bed of particulate solids is preheated to ~1000°F by passing heated air through the bed and using heaters mounted on the reactor wall. Coal is then introduced into the bed and ignited, increasing the bed temperature to the desired operating temperature (e.g., ~1600°F). The bed is maintained at a selected temperature by passing air or an air-H_2O mixture through annular chambers on the exterior of the combustor wall.

In one-stage combustion experiments (with an oxidizing atmosphere in the bed), all combustion air is introduced at or near the bottom of the fluidized bed.

In two-stage combustion, a stoichiometric deficiency of air is introduced at the bottom of the bed to partially burn the fuel. All or most of the oxygen in the air fed to the first stage is consumed, and reducing conditions prevail in the bed. In some ANL experiments, additional air was introduced into the freeboard above the bed through a tube located about 6 inches above the fluidized bed; oxygen in this air feed reacted with the CO, hydrocarbons, and the unburned carbon elutriated from the bed.

The coals used in the various series of experiments were : (1) Illinois coal from Seam 6, Peabody Coal Co. Mine 10, Christian County, Illinois (furnished by Commonwealth Edison); and (2) Pittsburgh Seam Coal from the Humphrey Preparation Plant, Osage, West Virginia. Sulfur contents of the coals (on an as-received basis) were 3.7 and 2.4 weight percent respectively. The as-received coal was crushed to pass a -14 mesh sieve whereupon more than 80 percent of the coal was in the -14, + 170 mesh fraction. Limited additional size reduction occurred as the coal passed through the screw feeder.

A residual crude oil was obtained from Esso Research and Engineering Co. Its sulfur content was 1.9 weight percent, viscosity (Seconds Saybolt Furol) was 162.5, and the flash point was 178°C.

The natural gas (obtained from Northern Illinois Gas Company) had a heat content of 1035 Btu/ft^3.

The additive materials studied include: (1) limestone No. 1359, Stephen City, Virginia (97.8 wt % $CaCO_3$, 1.3 wt % $MgCO_3$); (2) limestone No. 1360, Monmouth, Illinois (69.8 wt % $CaCO_3$, 19.2 wt % $MgCO_3$); (3) dolomite

No. 1337, Gibsonburg, Ohio (53.4 wt % $CaCO_3$, 46.5 wt % $MgCO_3$); and (4) Tymochtee dolomite, Huntsville, Ohio (49.3 wt % $CaCO_3$, 36.6 wt % $MgCO_3$).

Results and Discussion

1. Air Excess, Coal Combustion Experiments

The first experiments discussed here were made with sufficient fluidizing air introduced at the bottom of the reactor so that the gas leaving the fluidized bed contained unreacted oxygen. A large-particle-size limestone was fed, little of which was elutriated from the bed (which consisted of calcined, partially sulfated lime).

a. Effect of operating variables on SO_2 retention — For this mode of operation, the operating variables having significant effects on sulfur retention were the Ca/S mole ratio in the feed streams (the ratio of moles of calcium in the additive to moles of sulfur in the coal), the fluidized-bed temperature, and the superficial gas velocity. (Sulfur retention is defined as the percentage of the sulfur associated with the coal feed that is not contained in the off-gas as SO_2.) Less significant variables were the types of coal and additive, the particle sizes of coal and additive, the height of the fluidized bed, and the amount of excess air fed to the fluidized bed. Three variables having no demonstrable effect on sulfur retention were: (1) premixing of coal and additive before they were fed to the combustor (instead of feeding separate streams of coal and additive), (2) temperature of the gas in the freeboard above the fluidized bed, and (3) addition of small quantities of water to the fluidized-bed.

Ca/S mole ratio. Figure 2 shows the effect of Ca/S mole ratio on sulfur retention at 1450°F for Pittsburgh coal and at 1550 and 1600°F for Illinois coal. Sulfur retention increased as additive (Ca) feed rate was increased in relation to the coal (S) feed rate. Relatively good removals were attained at Ca/S ratios above 3.

Fluidized-bed temperature. Experimental results indicate that there is a temperature at which sulfur retention is at a maximum over the range of bed temperatures studied (Figure 3). With Illinois coal, a Ca/S mole ratio of ~2.5, and limestone No. 1359 additive, the optimum bed temperature apparently was 1500-1550°F. With Pittsburgh coal, a Ca/S mole ratio of ~4.0, and the same limestone additive, 1450-1470°F appears to be the optimum temperature. Those results were obtained in experiments using a gas velocity of 3 ft/sec and 3 percent excess O_2 in the flue gas.

The difference in optimum temperatures may be associated with different properties of the coal or alternatively, the temperature for optimum sulfur retention may have been influenced by Ca/S mole ratio. In any case, an operating temperature of 1500°F would be near optimum.

Superficial gas velocity. Sulfur retention was observed to increase with decreased superficial gas velocity (in the range of 3.5 to 7.4 ft/sec) at a coal combustion temperature of 1550°F and with addition of limestone No. 1359 (> 1000 μm average particle size) and Illinois coal at a Ca/S mole feed ratio of ~4 (Figure 4). The relatively coarse additive was selected to ensure that additive particles would be retained in the fluidized bed at high gas velocities. At gas velocities of 3.5, 5.5, and 7.4 ft/sec, the average SO_2 concentrations in the flue gas were 770, 1250, and 1500 ppm, corresponding to retentions of 83, 73, and 66 percent of the sulfur fed to the reactor. These data may be correlated with the equation

$$R = 101.79\, e^{-0.0625\, v} \qquad (1)$$

where: R = SO_2 retention, %
v = superficial gas velocity, ft/sec

Results of British experiments (1) using Welbeck coal, 440-μm British limestone, Ca/S mole ratios of 1 and 2, and a coal-ash fluidized bed show that sulfur retention is greater at a gas velocity of 2 ft/sec than at 3 ft/sec (Figure 4). The slopes decrease as the Ca/S mole ratio increases; thus, at sufficiently high Ca/S ratios, sulfur retention may be essentially independent of superficial gas velocity.

Excess air. The oxygen level in the off-gas was varied by adding pure oxygen at several rates to the fluidizing air before it entered the preheater. At 1550°F, ~3 ft/sec gas velocity, and Ca/S mole ratio of ~3, the sulfur retentions were 67, 71, and 75 percent, respectively, at 0.7, 2.4, and 5.6 percent O_2 in the flue gas. Apparently, oxygen concentration affects slightly the reaction of SO_2 with limestone, and sulfur retention can be expected to increase when oxygen concentration in the flue gas is increased.

Fluidized-bed height. Runs were performed at 1550°F, 3 ft/sec gas velocity, and Ca/S of ~4 with three different bed heights — 14, 24, and 46 inches (length to diameter (L/D) ratios of 2.3, 4.0, and 7.7). The sulfur retentions were 78, 80, and 83 percent, respectively, indicating that bed height has a small but real effect.

Type of coal. The effect of type of coal on sulfur retention could not be evaluated because of insufficient data in ANL experiments. However, qualitatively, sulfur retentions for Illinois and Pittsburgh coals differed little. Work by the British and by the U.S. Bureau of Mines has evaluated this variable in greater detail.

Type of additive. Sulfur retentions for runs performed at a Ca/S mole ratio of 2.5 and fluidized-bed temperatures of 1550 or 1600°F with several types of additives may be compared in Figure 2. Sulfur retention varied about 10 percent indicating that differences in these additive types had only small effects at these operating conditions.

Size of coal particles. To determine the effect of sulfur retention of the particle size of the coal feed, two experiments were completed with -12 +50 mesh and -50 mesh (a -12 +50 fraction ground to -50 mesh) Illinois coal at a Ca/S mole feed ratio of 2.4 and a fluidized-bed temperature of 1550°F. Sulfur retentions by No. 1359 limestone additive were 81 and 75 percent, respectively, for the -12 +50 mesh and -50 mesh feeds. An experiment in which Illinois -14 mesh coal was burned under operating conditions nearly identical to those used in the above experiments yielded a 78 percent sulfur retention. Thus, similar sulfur retentions were obtained by burning coal of three particle size distributions, but the coarsest coal feed appears to yield the best results.

Size of additive particles. Sulfur retention was calculated by interpolation to be ~87 percent for larger additive particles (1000 μm average) and 93 percent for smaller particles (630 μm average) in runs with a Ca/S mole ratio of 4.0, a temperature of 1550°F, and a gas velocity of 3 ft/sec. This suggests that at least in the region of high sulfur retention, additive particle size in this range has only a moderate effect on sulfur retention.

b. NO levels in the flue gas — Nitrogen oxides, principally nitric oxide (NO), are formed during the combustion of fossil fuels and are an important contributor to air pollution. Although the quantities of NO observed in the flue gas from high-temperature conventional combustors may be accounted for by the equilibrium of the nitrogen fixation reaction, this is not the case for low-temperature fluidized-bed coal combustion in which NO concentrations far in excess of those expected on the basis of the equilibrium have been observed. At 1600°F, a common temperature for fluidized-bed combustion, the equilibrium concentration of NO from the fixation of atmospheric nitrogen ranges from 50 to 200 ppm, depending on the oxygen concentration. (The oxygen concentration, in turn, depends on the level of excess air employed.) However, in actual fluid-bed coal combustion, nitric oxide levels of 400 to 800 ppm in the flue gas have been measured with oxygen concentrations of ~3 volume percent in the off-gas (15 to 20 percent excess air).

In previous ANL work, the major source of nitric oxide during the combustion of coal was determined to be the nitrogenous content of the coal (1 to 1.5 weight percent in U.S. coals). In more recent work, the effect of moisture content of the coal on NO level was studied by adding water at several rates to the fluidizing air. Pittsburgh coal and limestone No. 1359 were the feed materials. The fluidized-bed temperature was 1450°F and the Ca/S mole ratio was 1. The concentration of NO decreased from 530 ppm to 510 ppm when 10 cm^3/min water was added (equivalent to 26 weight percent water in the coal), and to 380 ppm when the rate of water addition was further increased to 30 cm^3/min (equivalent to 51 weight percent water in the coal). These decreases in NO concentration may be due to chemical reduction of NO by hydrogen produced by the water-gas-shift reaction, but the effect is not great enough to warrant further attention.

c. Calcium utilization — The relative extent of conversion of CaO to $CaSO_4$ for bed and elutriated materials was calculated from calcium and sulfur concentrations determined by wet chemical analysis. In several experiments with No. 1359 limestone of an average particle size of 490 μm, at temperatures ranging from 1400 to 1600°F, and gas velocities from 2.5 to 2.8 ft/sec, the conversion of calcium oxide to calcium sulfate was highest (2/3 converted) for particles collected on the final filter. These particles have a high surface to volume ratio and would be expected to react rapidly even though their residence time in the bed is relatively short. Next highest conversion, ~2/5, was obtained in bed particles which have a relatively long residence time in the bed. Lowest conversion, ~1/4, was obtained with solids removed from cyclones. These particles are larger than final filter particles, smaller than the bed particles, and are present in the bed for a relatively short time.

2. Air Deficient, Coal Combustion Experiments

The concept of two-stage combustion provides for a substoichiometric quantity of air (that is, less air than is required to burn the coal completely to CO_2 and H_2O) introduced into the first stage of the fluidized bed to which coal is fed. Additional air may be injected into the disengaging section above the fluidized bed (the second stage) to burn gaseous hydrocarbons, H_2, and CO in the gas stream from the first stage.

Two-stage combustion experiments of an exploratory nature were conducted to determine if this combustion mode might have benefits, as compared with single-stage fluidized-bed combustion. To simulate the conditions of combustion in the first stage only, experiments were performed in which a substoichiometric quantity of air was introduced into the bottom of the fluidized bed, but no secondary air was fed. In other experiments, secondary air was introduced above the fluidized bed. The bed consisted of coarse lime particles in all of these experiments.

The experimental results include information on the concentrations of SO_2, H_2S, NO, and CO in the off-gases at various air feed rates and bed temperatures, as well as data on the sulfur content of solid products when a substoichiometric quantity of air was fed to the first stage only.

a. Effect of decreasing air input on ratio of H_2S to $H_2S + SO_2$ in flue gas — The concentration of H_2S in the off-gases was measured to determine which operating conditions affect the formation of this sulfur compound. In experiments in which air was introduced to the first stage only, the amounts of H_2S and SO_2 in the off-gas were compared. The percentage of sulfur in the off-gas as H_2S was sensitive to the amount of air introduced into the fluidized bed, increasing drastically when the air feed rate was reduced below a value corresponding to ~70 percent of the stoichiometric quantity of air necessary to react with the coal feed (see Figure 5). (Although the parameter, air feed rate as a percent of stoichiometric, was based on feed rates of coal and air, it is recognized that the quantity of coal actually oxidized varies with other parameters (i.e., temperature, etc.). For certain correlations, it may well be more suitable to use the parameter, stoichiometric air feed rate based on the coal actually oxidized.) At an air feed rate equivalent to ~50 percent of the stoichiometric quantity, the concentration of H_2S (611 ppm) was nearly equivalent to the concentration of SO_2 (660 ppm). At air inputs of 70 to 80 percent of the stoichiometric quantity, the relative amount of sulfur as H_2S fell to about 2 percent of the total sulfur in the gas.

In those experiments in which secondary air was introduced above the fluidized bed (Figure 5), the H_2S level in the off-gas was low — corresponding to less than 1 percent of the total sulfur in the gas. This suggests that any H_2S in the gas leaving the first stage is oxidized to SO_2 by air fed to the second stage.

No consistent relationship was apparent between H_2S level and either the temperature of the fluidized bed (1450-1650°F) or the temperature of the off-gas in the freeboard above the bed (1100-1800°F).

b. Sulfur retention — Sulfur retention is defined as the percentage of the sulfur associated with the coal feed but not contained in the off-gas as SO_2 or H_2S. (Because a fraction of the carbon was not burned in these experiments, the sulfur retention values given are probably higher than would be realized if all of the carbon were burned.) Experiments were performed with no introduction of secondary air (Figure 6) to determine sulfur retention as a function of the Ca/S mole ratio in the feed at 1450, 1550, and 1650°F. Also shown in Figure 6 (to allow comparison) are data for experiments carried out earlier under one-stage oxygen-excess conditions at 1450-1470°F.

The data presented for the sub-stoichiometric air experiments show a large amount of scatter principally due to variation in the quantity of air fed to the fluidized bed, which was not the same in all experiments. Stoichiometric air added in each experiment ranged from 51 to 91 percent. The best retentions were observed at stoichiometric additions of less than 60 percent.

For experiments carried out at a Ca/S ratio of about 2 and temperatures of 1450-1650°F (Figure 7), no simple relationship between the amount of air introduced into the bed and sulfur retention was evident; however, a line has been fitted to the points as shown. At an air feed rate of 100 percent of stoichiometric, sulfur retention is about 65 percent. As the air feed rate is decreased, sulfur retention first decreases to about 45 percent as the air rate is decreased to 75 percent of the calculated stoichiometric requirement and then increases rapidly as the air rate is decreased further. This suggests that in an oxygen-deficient region (75 to 95 percent of calculated stoichiometry), removal of sulfur by lime in the form of SO_2 is poor, but that at lower air flow rates sulfur is in the form of H_2S and is efficiently removed. This would be expected because oxidizing conditions are required for the retention of SO_2 by lime (to convert a $CaSO_3$ intermediate to $CaSO_4$), whereas reducing conditions are required for the retention of H_2S by lime.

The introduction of secondary air above the bed resulted in erratic but generally lower sulfur retentions. Decreases were about 10 percent at 1450°F, 20 percent at 1550°F, and 40 percent at 1650°F. The increased sulfur content of the off-gas after secondary air was introduced was probably caused by burning of entrained coal particles in the second stage to produce additional SO_2.

c. NO levels in the flue gas — When coal was burned with a deficiency of air fed to the first stage, concentrations of NO in the off-gas from the first stage as a function of the amount of air introduced into the bottom of the fluidized bed were as shown in Figure 8. To obtain these data, only the first stage was operated. The NO concentrations were generally < 250 ppm and apparently were affected by both the amount of air introduced and the temperature of the fluidized bed. At the lower air feed rates, the NO levels were generally lower. At a given air feed rate, NO levels were higher at lower bed temperatures. Since earlier work at ANL showed that nitrogenous compounds in coal are oxidized to NO during fluidized-bed combustion, it can be postulated that the lower levels of NO observed at higher temperature are due to more rapid decomposition of NO. This decomposition may be promoted by the presence of CO; another possibility is that nitrogenous compounds other than NO may be formed in the highly reducing atmosphere of the bed.

The data presented in Figure 8 are not corrected to an equivalent off-gas volume basis. However, if this correction were made, the dependence of NO emissions on air feed rate would be even more pronounced, assuming that feed rates of coal were equivalent.

Upon the introduction of secondary air above the fluidized bed, NO levels in the off-gas varied eratically — usually increasing. Possible explanations for this behavior are: (1) any reduction of NO by CO in the zone above the bed would be suppressed by introducing secondary air or (2) if a nitrogen compound such as ammonia were present in the gas, it may be oxidized to NO by the secondary air.

d. Sulfur species in the bed — Results show that the sulfide content of bed material decreased as air flow was increased. At air inputs corresponding to 50 percent of stoichiometric, as much as 100 percent of the sulfur in the bed was sulfide. In most experiments in which the air input exceeded 65 percent of stoichiometric, sulfide content dropped off rapidly to less than 1 percent. No relationship was found between sulfide content and bed temperature.

The sulfite content of bed samples was erratic, ranging between 6.2 and < 0.1 weight percent. No correlation of sulfite content with either bed temperature or air feed rate could be found.

e. Carbon balance — Carbon balances were made for three experiments. For three other experiments, all data for making the balances except the CO_2 level in the flue gas are available (see Table 1). The small quantity of carbon not accounted for is represented by hydrocarbons (other than CH_4) for which no analyses are made and by a small loss of fine carbon particulate from the combustion system. The data show that as the volume of air added to the bed decreases (experiments 14-1A, -1B, -2) at the same temperature, the CO content of the flue gas increases markedly, the CH_4 content increases slightly, and the quantity of carbon elutriated to the first and second cyclone separators from the fluidized bed increases.

The carbon content of the bed under low stoichiometric air additions (55 percent) was as high as 31 percent (experiment 14-3B). Under these conditions the amount of carbon elutriated was about 15 percent of that fed.

f. Preliminary evaluation of the concept — Although the work conducted on two-stage combustion was exploratory in nature, a preliminary evaluation of the concept can be made. The principal advantages of the two-stage combustion concept over one-stage combustion are: (1) lower NO emissions; (2) retention of sulfur in the form of calcium sulfide (rather than sulfate), allowing for potentially easier regeneration of the additive; and (3) production of a combustible gas that could be used in conjunction with a gas turbine.

The principal disadvantages are: (1) greater elutriation of carbon, (2) possible complications in additive regeneration owing to the high carbon content of the bed, and (3) necessity for removal of heat from the bed under conditions that might be corrosive to immersed steam tubes.

Table 1. CARBON BALANCES AND CARBON CONTENTS OF SOLIDS IN SUBSTOICHIOMETRIC AIR-COMBUSTION EXPERIMENTS

	14-1A	14-1B	14-2	14-1C	14-3A	14-3B
Carbon in coal, g/hr	1428	1428	1684	1799	1485	1713
Carbon out, g/hr						
First cyclone	71	136	174	167	235	233
Second cyclone	22	11	24	7	7	16
Flue gas						
CH_4	>24	32	39	42	28	32
CO	42	350	441	521	330	455
CO_2	1230	ND[a]	840	ND	ND	800
Total carbon out, g/hr	>1470	ND	1632	ND	ND	1682
Carbon concentration in solids streams, wt%						
Bed	<1	<1	6	<1	20	31
First cyclone	29	39	46	39	47	62
Second cyclone	52	53	55	56	35	53
Run conditions						
Temperature, °F	1450	1450	1450	1550	1600	1600
Coal feed, lb/hr	5.0	5.0	6.3	5.9	5.2	6.0
Air, % of stoichiometric	90	86	71	54	64	55

[a] No data available.

Sulfur retention appears to be roughly equal for the two concepts. It is notable, also, that no problems of coal caking were encountered even at high bed carbon contents.

Further work might be warranted at lower air addition rates and higher bed temperatures to avoid heat generation in the bed. Under such conditions the process would become gasification rather than combustion.

3. Air Excess, Oil Combustion Experiments

To assess the removal of SO_2 from combustion gases when residual fuel oil is burned in a fluidized bed of sulfated lime with continuous feeding of limestone additive, experiments were performed in the 6-in. diameter fluidized-bed combustor at a variety of operating conditions. Residual fuel oil was burned in an excess of oxygen at bed temperatures ranging from 1450 to 1650°F, Ca/S mole ratios up to 11.9, a gas velocity of ~3 ft/sec (except for one experiment at 5.5 ft/sec), and with 3 volume percent oxygen in the flue gas (except in one experiment with 1 volume percent oxygen in the flue gas).

The following results were obtained in these experiments.

1. The effect of temperature on sulfur retention is similar to that observed in coal combustion experiments, in which there is a temperature yielding maximum sulfur retention. In the oil-combustion experiments, maximum sulfur retention was at 1500-1550°F.

2. The shape of the curve for sulfur retention as a function of Ca/S mole ratio is similar to that obtained in coal combustion experiments. Sulfur retention in the oil combustion runs increases as Ca/S mole ratio increases to about 5, then levels off at a 90 percent sulfur retention level as the Ca/S ratio is increased further. The slope of the curve for sulfur retention as a function of Ca/S ratio is less steep than the slope for Illinois coal (3.7 weight percent sulfur) at similar operating conditions.

When oil was combusted, the NO levels in the flue gas ranged from 110 to 150 ppm for the experiments with 3 percent O_2 in the flue gas. This may be compared with the 400 to 800 ppm range observed when coal was burned. However, the nitrogenous content of residual oil is also less than that of coal. No correlation of NO level with bed temperature or Ca/S mole ratio was observed. Combustion efficiency in these experiments is discussed in the section of combustion efficiencies below.

4. Miscellaneous

a. Additive decrepitation rates during coal combustion experiments — Decrepitation and attrition of several additives during coal combustion experiments has been estimated from the calcium content of elutriated fines. (The fraction of additive carried over can only be estimated because the particle matter elutriated during the combustion of coal in a fluidized bed is a mixture of solids of different origins and compositions.)

In most experiments a gas velocity of ~2.6 ft/sec was used; at this velocity all of the flyash and additive particles having diameters of <177 μm are expected to elutriate from the fluidized bed.

The expected elutriation for each of several series of experiments calculated in this manner is shown in Table 2. The actual elutriation was determined from calcium material balances (with an allowance made for the calcium content of the flyash, which was also expected to elutriate). The difference between actual and expected elutriation gave an estimate for the decrepitation of larger additive particles (Table 2).

The indicated decrepitation of BCR-1359 limestone was ~8 percent, but no decrepitation of a British limestone was evident. Decrepitation of limestone BCR-1360 and dolomite BCR-1337 was more severe—40 and 85 percent, respectively. These results indicate that decrepitation of BCR-1359 and British limestone is low and that limestones of this

Table 2. ESTIMATED DECREPITATION OF ADDITIVE MATERIALS FROM FLUIDIZED-BED COMBUSTOR

Experiments	Additive type	Distribution of total calcium in combustor system, wt %		
		Expected elutriation[a]	Actual elutriation[b]	Estimated decrepitation
Amer-1,-3,-4	BCR-1359	12	19	7
BC-6,-7,-8	BCR-1359	2	11	9
AR-1,-2,-4,-5,-6	BCR-1359	13	21	8
BC-9	BCR-1360	2	42	40
BC-10	BCR-1337	< 2	~ 87	85[c]
Brit-1,-2,-3,-3A and Amer-Brit	British limestone	37	33	—

[a] Calcium contained in particles $< 177 \mu m$ diameter in the additive feed to the system. These particles are expected to elutriate at a superficial velocity of 2.6 ft/sec. Fine particles in the starting fluidized bed are not included.

[b] Calcium fed with the coal was deducted from the total calcium found in the elutriated material.

[c] Derived from both calcium and magnesium material balances.

type are desirable materials for use in a full-scale fluidized-bed combustor with regeneration and recycle of additive. Higher decrepitation rates for BCR-1360 and BCR-1337 may make these materials less promising for regeneration and recycle. Data are for one cycle of use only.

b. Cyclone collection efficiencies during coal combustion experiments — Data on the particle removal efficiency of the ANL cyclone separators have been compiled as a basis for estimating the dust loading and filter area of a cartridge filter for a pressurized combustion bench-scale plant now being designed. In the present atmospheric pressure system, flue gas passes through two cyclones in series and a final filter. (The diameter of the first in-line cyclone is 6-5/8 inches; the diameter of the second is 4-1/2 inches.) It is planned to use the same two cyclones with the pressurized combustor.

To determine the adequacy of the glass fiber mat filters used in the atmospheric plant, collection efficiencies (defined as ratio of the weight of particles removed to the weight of particles entering the cyclone) were compiled for 26 earlier ANL experiments. In these one- and two-stage runs, the flue gas flowrates ranged from 8 to 14 ft^3/min, the coal feed rates from 4 to 7.3 lb/hr, the additive feed rates from 1.1 to 2.3 lb/hr, and the dust loadings at the combustor exit from 0.16 to 1.78 g/ft^3. Combined efficiency of the two cyclones was above 80 percent in 24 of the 26 experiments and above 90 percent in 21 of the experiments. The dust loading in the flue gas leaving the second cyclone averaged 0.06 g/ft^3 for the 26 runs; the maximum loading was 0.22 g/ft^3.

c. Combustion efficiencies for coal, oil and gas with excess air — The combustion efficiency for experiments performed in the combustor was determined as the ratio of carbon burned to carbon fed, multiplied by 100. The carbon loss is calculated by determining unburnt carbon leaving the system by three routes: (1) carbon associated with the elutriated solids; (2) incompletely burned gases (e.g., carbon monoxide and hydrocarbons); and (3) carbon associated with fluidized-bed material taken from the system. All experiments were conducted without recycle of fines.

Combustion efficiencies in ten experiments with coal ranged from 93 to 96 percent. In all experiments, carbon losses in the bed material were negligible. Only about 10 to 20 percent of the carbon loss was due to the formation of carbon monoxide and hydrocarbons. The major carbon loss (80 to 90 percent) occurred as a result of elutriation of fine particles in the exhaust gases before they were completely combusted. Combustion efficiency can be increased by recycling the elutriated ash-carbon mixture to the fluidized bed or to a carbon burnup cell. Oxygen concentration in the flue gas in these experiments was approximately 3 percent.

Combustion efficiency in oil combustion experiments was similar to that observed for coal combustion experiments under similar conditions, ranging from 94 to 96 percent for experiments with 3 percent O_2 in the flue gas. However, sources of carbon losses for the two fuels differed. In coal combustion, most of the carbon loss is represented by the carbon content of solids elutriated to the cyclones; in oil combustion, inefficiency results from incomplete burning of the CO and hydrocarbons formed during combustion. Efficiency can probably be improved by operating the combustor with a deeper bed or by increasing the freeboard temperature. Lower combustion efficiencies were observed with less excess oxygen in the flue gas and at higher gas velocities.

During the combustion of natural gas, the elutriation of carbon-bearing fine particles is negligible, and the major loss of unburnt carbon is in the carbon monoxide and total hydrocarbons in the flue gas. Combustion efficiencies, calculated from analyses of samples of the flue gas, ranged from 94.1 to 99.2 percent at 1600°F with 3 percent excess oxygen. At 1800°F, combustion efficiencies of 94.1 to 98.8 percent were observed. These results are similar to data reported by the USSR on combustion of gas in a fluidized bed. Although the USSR data indicate that combustion efficiency is principally affected by bed temperature, combustion efficiency is also likely to be a function of bed depth, gas velocity, and excess oxygen concentration. For example, in experiment NG-3 at 1800°F, combustion efficiency was decreased to 91 percent when the combustion was intentionally forced toward more reducing conditions; i.e., 1.5 volume percent O_2 in the flue gas rather than 1.8-4.5 volume percent O_2.

REGENERATION OF SULFUR CONTAINING ADDITIVES

When coal is burned in a fluidized bed containing limestone or dolomite, sulfur-containing gases from the combustion of sulfur-containing substances in the coal react with the bed material and are retained in the bed. The reaction product is calcium sulfate if combustion is carried out under oxidizing conditions, or calcium sulfide if combustion is carried out with a deficiency of air.

Several regeneration processes are under consideration. These are:

1. Reductive decomposition of calcium sulfate.
2. Roasting of calcium sulfide in air or oxygen.
3. Reaction of calcium sulfide with water and carbon dioxide.

Processes 2 and 3 can be used to regenerate not only calcium sulfide, but also material containing calcium sulfate if the sulfate is first reduced to the sulfide.

Thermodynamic Analyses

Valuable information can be gained by considering the thermodynamics of the process. The yields of gaseous sulfur-containing products, the composition of solid phases, and the variations of these yields and compositions with temperature, pressure, and gas composition for a system at equilibrium can all be obtained. Optimum reactant feed ratios and gas compositions can also be calculated easily when product concentrations, compositions, and pressures are specified.

All of the following predictions and conclusions are based on the supposition that chemical equilibrium is achieved among the various phases. This implies that the rates of all relevant chemical reactions are large on the time scales being used, which scales are determined by mass transport rates within the system. The maximum rates at which this supposition is valid vary with temperature and must be determined in the laboratory and in the pilot plant. It is further assumed that the system is not stoichiometrically limited. There must always be at least small amounts of the appropriate solid phases present for the results of these calculations to be valid. One must not assume that all actual processes will be operated with all these solid phases present, however.

The assumption is also made that solid solutions do not form to any great extent. Exploratory experiments to date support this assumption.

1. Reductive Decomposition of $CaSO_4$ with CO/CO_2

Before the relative amounts of the species in an equilibrium mixture from the reduction of calcium sulfate with carbon monoxide–carbon dioxide mixtures can be calculated, the solid phases present at the various conditions of temperature and carbon monoxide/carbon dioxide ratio must be determined. The possible sulfur-containing solids are considered to be calcium sulfate, calcium sulfite, and calcium sulfide.

a. Conditions for the presence of calcium sulfate and calcium sulfide — The solid phases present at equilibrium with a P_{CO}/P_{CO_2} of 0.005-0.055 and temperatures of 1600 to 2400°F are shown in Figure 9 (in which temperature is the ordinate and P_{CO}/P_{CO_2} the abscissa). Examination of the expression for K_p in reaction 2

$$1/4\ CaSO_4 + CO \rightleftharpoons 1/4\ CaS + CO_2$$

$$K_p = \frac{P_{CO_2}}{P_{CO}} \tag{2}$$

shows that for any temperature, there is but one ratio of carbon monoxide to carbon dioxide at which calcium sulfate and calcium sulfide can coexist at equilibrium. The coexistence conditions appear as the line running from the lower left to the upper right part of Figure 9 and represent CO/CO_2 ratios at which CaS and $CaSO_4$ can both be present at equilibrium. In the area to the right of this line, the gas mixture is so rich in carbon monoxide that calcium sulfate is completely reduced to calcium sulfide. To the left of the line, the gas mixture is so rich in carbon dioxide that calcium sulfide is completely oxidized to calcium sulfate. This line is called the coexistence line for calcium sulfate and calcium sulfide.

b. Conditions for presence of calcium sulfite — Calcium sulfite is not stable in the presence of CO/CO_2 mixtures at any temperature from 1500 to 2400°F. This has been established by plotting "coexistence" lines for calcium sulfite with calcium sulfate and for calcium sulfite with calcium sulfide. These are analogous to the calcium sulfate-calcium sulfite coexistence line described above and are determined in the same way from the equilibrium constants for reactions 3 and 4.

$$CaSO_4 + CO \rightleftharpoons CaSO_3 + CO_2$$

$$K_p = \frac{P_{CO_2}}{P_{CO}} \tag{3}$$

$$1/3\,CaS + CO_2 \rightleftharpoons 1/3\,CaSO_3 + CO$$

$$K_p = \frac{P_{CO}}{P_{CO_2}} \qquad (4)$$

c. Conditions for the presence of calcium carbonate and calcium oxide — When calcium sulfate is reduced or calcium sulfide is oxidized by a mixture of CO and CO_2, calcium oxide is formed. However, in the presence of carbon dioxide at sufficient pressure, calcium oxide is converted to calcium carbonate.

A coexistence line for the carbonate and the oxide is determined by the equilibrium dissociation pressure of calcium carbonate (reaction 5).

$$CaCO_3 \rightleftharpoons CaO + CO_2$$

$$K_p = P_{CO_2} \qquad (5)$$

It also appears as a nearly horizontal line at about 1950° F in Figure 9. This line represents the temperature at which the partial pressure of CO_2 in the equilibrium mixture just equals the equilibrium pressure of CO_2 over calcium carbonate.

The partial pressure of CO_2 in the equilibrium mixture is obtained by assuming a total pressure of 10 atm and subtracting the pressures of SO_2 and CO. Clearly, if the total pressure is lowered or if an inert gas is added, the pressure of CO_2 will be lower and the horizontal line will be at a lower temperature. It is also clear that the calcium carbonate does not exist above the horizontal line and that calcium oxide does not exist below it.

d. Sulfur dioxide pressure -- The pressure of sulfur dioxide in the equilibrium mixture can be calculated from the CO/CO_2 ratio and the equilibrium constant of the reaction appropriate to the part of Figure 9 under consideration; however, in the areas labeled C and D, one must generate independent information about the CO_2 pressure by making assumptions exactly analogous to those made in the above discussion of calcium carbonate.

In area A of Figure 9, SO_2 is generated by reaction 6.

$$CaSO_4 + CO \rightleftharpoons CaO + SO_2 + CO_2$$

$$K_p = \frac{P_{SO_2} \cdot P_{CO_2}}{P_{CO}} \qquad (6)$$

The pressure of SO_2 is shown as a family of isobars slanted down toward the right. In area B, SO_2 is generated by the oxidation of calcium sulfide in accordance with reaction 7.

$$1/3\,CaS + CO_2 \rightleftharpoons 1/3\,CaO + CO + 1/3\,SO_2$$

$$K_p = \frac{\left(P_{SO_2}\right)^{1/3} P_{CO}}{P_{CO_2}} \qquad (7)$$

The isobars of constant SO_2 pressure in area B curve down to the left, meeting those of area A at the calcium sulfide-calcium sulfate coexistence line. At any temperature, the SO_2 pressure is at a maximum at this junction. For example, at 2000° F, the maximum attainable equilibrium pressure of SO_2 is 0.46 atm at a CO/CO_2 ratio of 0.020. This maximum in the SO_2 pressure may be understood by examination of the appropriate equilibrium constants. For example, the expression for the equilibrium constant for reaction 6 predicts that the pressure of SO_2 is directly proportional to the CO/CO_2 ratio. Thus, the pressure of SO_2 must increase as the CO/CO_2 ratio increases as long as reaction 6 obtains. From the expression for the equilibrium constant in reaction 7, it may be seen that the SO_2 pressure is inversely proportional to the cube of the CO/CO_2 ratio. Thus, the SO_2 pressure increases with decreasing CO/CO_2 ratio as long as reaction 7 obtains. Reactions 6 and 7 occur simultaneously only along the coexistence line. Thus, as one moves away from the coexistence line, the SO_2 pressure must decrease.

In area C, reaction 8 applies.

$$CaSO_4 + CO \rightleftharpoons CaCO_3 + SO_2$$

$$K_p = \frac{P_{SO_2}}{P_{CO}} \quad (8)$$

In this area, the pressure of SO_2 is dependent only on K_p and carbon monoxide pressure, and the SO_2 isobars are nearly vertical. The main effect results from the variation of K_p with the temperature. In area D, the SO_2 pressure is once again a strong function of the CO/CO_2 ratio as may be seen from reaction 9.

$$CaS + 4\,CO_2 \rightleftharpoons CaCO_3 + SO_2 + 3\,CO$$

$$K_p = \frac{P_{SO_2}\left(P_{CO}\right)^3}{\left(P_{CO_2}\right)^4} \quad (9)$$

The slope of the isobars in area D differs only slightly from the slope in area B as a result of increased dependence on CO_2 pressure in area D.

e. Sulfur pressure — The pressure of sulfur vapor is quite low ($<10^{-2}$ atm) over the temperature range 1700 to 2300°F. The pressure of sulfur in area B was calculated from reaction 10.

$$CaS + CO_2 \rightleftharpoons 1/2\,S_2 + CO + CaO$$

$$K_p = \left(P_{S_2}\right)^{1/2}\frac{P_{CO}}{P_{CO_2}} \quad (10)$$

Since the formation of sulfur is entirely analogous to the formation of SO_2, sulfur concentration may be expected to exhibit the same sort of maximum at the calcium sulfide-calcium sulfate coexistence line.

f. Carbonyl sulfide pressure — The carbonyl sulfide pressure was calculated with reaction 11.

$$CaS + CO_2 \rightleftharpoons CaO + COS$$

$$K_p = \frac{P_{COS}}{P_{CO_2}} \quad (11)$$

Since P_{COS} is dependent on the pressure of CO_2, assumptions made in calculating COS pressure are similar to those made in the discussion of calcium carbonate. P_{COS} is low ($\sim 10^{-3}$atm) in this system.

g. Solid-solid reaction of calcium sulfide with calcium sulfate — SO_2 is generated by the reaction of calcium sulfide with calcium sulfate, as is shown in reaction 12.

$$1/3\,CaS + CaSO_4 \rightleftharpoons 4/3\,SO_2 + 4/3\,CaO$$

$$K_p = \left(P_{SO_2}\right)^{4/3} \quad (12)$$

Since reaction 12 is exactly equivalent to the sum of reactions 6 and 7, the SO_2 pressure calculated from reaction 12 must be just that calculated from reaction 6 or reaction 7 using the CO/CO_2 ratio at the coexistence line. Another way of saying this is that the presence of both calcium sulfide and calcium sulfate determines an oxidizing potential for the atmosphere with which it is in equilibrium; this oxidizing potential determines the CO/CO_2 ratio of the atmosphere. If carbon monoxide and carbon dioxide are present in the gas phase over a mixture of calcium sulfide and calcium sulfate, they serve as a facile route to the production of SO_2 so that rapid reaction rates for mixtures of the two solids are possible.

h. Reduction of calcium sulfate with H_2/H_2O mixtures — The system calcium sulfate-calcium sulfide-H_2-H_2O is exactly analogous to the system calcium sulfate-calcium sulfide-CO-CO_2. This means that all the features of the CO-CO_2 system are present in the H_2-H_2O system. The sulfur-containing solid phases once again are calcium sulfate and calcium sulfide, but calcium oxide and calcium hydroxide are the non-sulfur-containing solid phases. The pressure of SO_2 at a given temperature has a maximum value where the calcium sulfate and calcium sulfide are in equilibrium with the H_2-H_2O mixture. The instability of calcium sulfite can be shown in the same way as in the CO-CO_2 system. In fact, the major differences between the H_2-H_2O system and the CO-CO_2 system are that carbonyl sulfide is replaced with H_2S and that at any given temperature, the numerical value for the H_2/H_2O ratio differs from that of the CO/CO_2 ratio.

An additional difference between the systems is that in the H_2-H_2O system, calcium hydroxide can form at lower temperatures and higher pressures of H_2O (just as $CaCO_3$ can form in the CO-CO_2 system). However, at 10-atm H_2O pressure, $Ca(OH)_2$ is not stable above 1200° F.

For any temperature and SO_2 pressure, the H_2/H_2O ratio can be calculated from the equivalent CO/CO_2 ratio via reaction 13.

$$H_2 + CO_2 \rightleftharpoons H_2O + CO$$

$$K_p = \frac{P_{CO}/P_{CO_2}}{P_{H_2}/P_{H_2O}} \qquad (13)$$

This is the familiar water-gas shift reaction. The principle involved here is that equal SO_2 pressures are obtained in the H_2-H_2O system and the CO-CO_2 system when the oxidizing potentials of the atmospheres are the same; i.e., when the two atmospheres are in equilibrium with each other.

An important conclusion is that the maximum pressure of SO_2 from any system in which calcium sulfate is reduced or calcium sulfide is oxidized is the pressure of SO_2 observed along either the H_2-H_2O coexistence line or the CO-CO_2 coexistence line. The same is true for S_2 pressures. The basis for these rather far-reaching conclusions is that in any process involving a reduction of calcium sulfate, the SO_2 and S_2 pressures will increase with increasing reducing ability of the atmosphere until calcium sulfide is formed. At that point, increasing the reducing ability of the atmosphere no longer increases the amount of SO_2 or S_2 formed, but rather causes the calcium sulfate to be transformed into calcium sulfide. Similarly, in a process in which calcium sulfide is oxidized, the SO_2 and S_2 pressures increase with increasing oxidizing ability of the atmosphere until calcium sulfate is formed. This point is again a limit, and SO_2 and S_2 pressures cannot be increased further.

2. Roasting of Calcium Sulfide

In the roasting process, calcium sulfide is oxidized with oxygen or air according to reaction 14.

$$CaS + 3/2\ O_2 \rightleftharpoons SO_2 + CaO$$

$$K_p = \frac{P_{SO_2}}{\left(P_{O_2}\right)^{3/2}} \qquad (14)$$

As may be seen by examining the equilibrium constant for reaction 14, the pressure of SO_2 at any temperature increases with increasing pressure of oxygen. However, it follows from the arguments presented above that above some definite oxygen pressure (given at any temperature by K_p of reaction 15),

$$1/2\, CaS + O_2 \rightarrow 1/2\, CaSO_4$$

$$K_p = \frac{1}{P_{O_2}} \qquad (15)$$

calcium sulfide is no longer stable, but is converted to calcium sulfate. At this particular oxygen pressure, the SO_2 pressure is that observed along the coexistence line in the CO-CO_2 system or in the H_2-H_2O system. Thus, what appear to be two very different processes, the reductive decomposition of calcium sulfate and the roasting of calcium sulfide, are in fact very similar. Both processes give rise to identical maximum SO_2 pressures at a given temperature.

3. Pressure Effects in the Above Processes

In all regeneration processes discussed above, the pressure of SO_2 is a function of the temperature or of the oxidizing ability of the atmosphere (in the case of the roasting and reductive decomposition processes). The pressure of SO_2 as a function of the total system pressure has not been discussed because SO_2 pressure is independent of the total system pressure in these processes. However, percent of SO_2 in the gas mixture is an inverse function of the total system pressure since the pressure of SO_2 is fixed at any temperature. The pressure of SO_2 is also independent of the presence of inert gaseous diluents if sufficient oxidizing or reducing gas is present. As stated above, in the case of reductive decomposition with CO-CO_2 mixtures, the presence of inert gases may affect the CO_2 pressure enough to change the reaction product from calcium carbonate to calcium oxide.

4. Acid-Base Reaction of Calcium Sulfide with H_2O and CO_2

Reaction 16 has been proposed as a regeneration reaction for calcium sulfide.

$$CaS + H_2O + CO_2 \rightleftharpoons CaCO_3 + H_2S$$

$$K_p = \frac{P_{H_2S}}{P_{CO_2} \cdot P_{H_2O}} \qquad (16)$$

Calcium sulfide is formed in the additive by: (1) burning coal in a fluidized bed of limestone or dolomite with a deficiency of air or by (2) reducing the $CaSO_4$ in the additive from a run in which combustion was with an excess of air. Unlike all other regeneration reactions discussed here, this reaction is pressure-sensitive. The percentage and the pressure of H_2S increase with increasing total system pressure. The pressure of H_2S is also sensitive to the presence of inert-gas diluents, in contrast to the previously mentioned regeneration schemes. The equilibrium constant for this exothermic reaction becomes smaller as the temperature is increased (Figure 10). This is in direct contrast to the other (endothermic) regeneration schemes mentioned.

Maximum H_2S yield is obtained when the H_2O/CO^2 ratio in the feed gas is 1 to 1, as may be seen by an examination of the equilibrium constant expression. Figure 11 shows the pressure of H_2S as a function of temperature, assuming 10-atm total pressure and an inlet gas stream composed only of H_2O and CO_2 at various ratios. However, H_2O may be readily removed from the product gas stream by condensation. Thus, higher values of H_2S concentration in a dried gas stream may be obtained by operating with an excess of H_2O in the inlet gas stream. For an inlet gas composition of 50 percent water and 50 percent CO_2 and temperatures of 1000-1400°F, Table 3 gives the percentage of H_2S in the gas effluent from the reactor and the percentage of H_2S in the same effluent after it has been dried.

Table 3. H_2S CONCENTRATION[a] IN DRIED AND UNDRIED PRODUCT GAS STREAM AT EQUILIBRIUM

Temperature, °F	% H_2S in undried gas	% H_2S in dried gas
1000	23.0	37.4
1100	11.4	20.5
1200	4.7	9.9
1300	2.7	5.1
1400	1.4	2.8

[a] Assumptions are 10-atm total pressure with 50 percent H_2O and 50 percent CO_2 inlet gas.

B. Experimental Studies

4. Reductive Decomposition of $CaSO_4$

To test the accuracy of the equilibrium compositions calculated for the reduction of $CaSO_4$ with CO/CO_2 mixtures, experiments have been performed in a static system. The apparatus consists of a horizontal tube reactor fabricated from recrystallized alumina. The tube is 36 inches long and has an ID of 3 inches with 1/4-in. walls. One end of the alumina reactor is closed and the opposite end is capped with a stainless steel 0-ring flange. The flanged end is outside the furnace.

The experiments are performed in the following manner. A 3-gram sample of $CaSO_4$ (Drierite) is placed in an alumina boat and loaded into the reactor. The system is closed and leak-checked. The $CaSO_4$ is dried at 500°F under vacuum for 15 to 20 hours. While the system is still at 500°F and isolated from the vacuum pump, a predetermined pressure (<1atm) of CO is added. The total pressure of the system is then increased to 1 atm by adding CO_2. The system is allowed to stand for 30 minutes; then a gas sample is taken through the septum, using a hypodermic syringe. The gas sample is analyzed for CO by gas chromatographic techniques.

Next, the pressure of the system is reduced to 400 mm Hg, and the temperature is increased to 1900°F, increasing the pressure to ~1 atm. With the system at 1900°F, samples of the gas mixture are obtained at selected intervals and are analyzed for CO and SO_2. After the final gas sample is obtained, the system is flushed with N_2 to remove reactant and product gases. The temperature is lowered to room temperature and the residue in the alumina boat is removed and analyzed for Ca, total S, and $S^{=}$ by wet chemical analytical techniques.

Experimental results presented in Figure 12 show good agreement, in most cases, between experimental and calculated values of SO_2 levels after reaction of $CaSO_4$ with CO/CO_2 over a range of ratios. The data also indicate that CO and SO_2 were being slowly removed by a secondary process. Analyses of the gas samples indicate that reaction was occurring in the ratio of one mole of SO_2 and one mole of CO to form one mole of CO_2 during the period of equilibrium at 1900°F (the other reaction product is probably elemental sulfur which was found on the cooler end of the alumina tube). This reaction was evident in those cases where the equilibrium time was very long (~20 hours) and/or the CO/CO_2 ratio was high. For these cases, the secondary reaction caused the points to fall off the line. In an experiment in which SO_2 was diluted with N_2 only, it was observed that SO_2 reacted with materials of construction to a small extent; SO_2 concentration decreased from 15 percent initially to 14 percent in 2 hours and to 13 percent in 20-1/2 hours at 1900°F.

2. Two-Step Process

A second regeneration procedure of interest is to first reduce the $CaSO_4$ to CaS and then to react the CaS with CO_2-H_2O to form $CaCO_3$ and H_2S. Experimental data on these process steps were obtained in a 2-in. diameter batch fluidized-bed reactor.

a. Conversion of $CaSO_4$ to CaS — The effect of temperature on the rate of conversion of $CaSO_4$ to CaS with H_2 was studied and is shown in Figures 13 and 14. The additive used was partially sulfated dolomite ($CaSO_4$/CaO/MgO), obtained in an earlier experiment in which the dolomite took up SO_2 during combustion of coal at 1550°F. In the H_2-reduction runs, the gas velocity was ~ 6 ft/sec and the system was at atmospheric pressure. Approximately three stoichiometric equivalents of hydrogen were added over the 5-hr period of the runs. As would be expected, the sulfide concentration in the bed material was lower at 1350°F than at either 1450°F or 1600°F at equivalent times. Percent conversions after 4.5 hours at 1350°F, 1450°F, and 1600°F were 18, 38, and 86 percent, respectively.

Since in actual practice CO may be the reducing gas, an additional experiment was performed with CO, and the percent reduction of $CaSO_4$ was compared with the expected reduction using H_2 (Figure 14). The CO compared favorably with H_2 at the temperature employed (1500°F); however, this may not be true at all temperatures.

b. Reaction of CaS with CO_2/H_2O — The product of each of the reduction experiments was carbonated at 10 atm to simulate a product that would be obtained in an actual 10 atm combustion-reduction experiment. This material was then reacted batchwise with an equimolar mixture of CO_2/H_2O at temperatures ranging from 900 to 1100°F, at a gas velocity of approximately 1 ft/sec and at 10 atm pressure in the 2-in. diameter fluidized-bed reactor. The H_2S concentration in the outlet gas was monitored using a quadrupole mass spectrometer.

The results to date have shown that:

1. The reaction producing H_2S is initially rapid, but the rate decreases after a short time. Typically, the reaction rate drops to near zero after several minutes.
2. The peak concentration of H_2S in the outlet gas is high and near the expected equilibrium value.
3. Typically, half or less of the CaS reacts.

In continuing work, the effects of process variables are being studied in an attempt to increase the quantity of CaS that is reacted.

C. Sulfation-Regeneration Cyclic Experiments

Since it will be desirable to reuse the additive material several times in commercial applications, a cyclic experiment has been performed to obtain data on the pickup and removal of sulfur from additive particles and to determine decrepitation and attrition of additive particles during sulfation-regeneration cycles. Six cycles of simulated combustion and two-stage regeneration were performed with a single bed of additive. The starting material (1.2 kg) was obtained from a coal combustion experiment in which dolomite No. 1337 had been used as additive. The initial sulfur content of the bed was 15.4 weight percent. The experiment was performed batchwise in the 2-in. diameter fluidized-bed reactor.

The sulfation portion of cycle 1 was omitted since the additive already contained sulfur. For the remaining cycles, the constituents of the sulfating gas were N_2, CO_2, H_2O, O_2, CO, and SO_2. The sulfation reaction was allowed to proceed until the bed material had essentially ceased further pickup of SO_2. After the bed had been sulfated, the $CaSO_4$ was converted to CaS, using H_2 or CO as reductant at 1550 to 1600°F and 10 atm. The bed was then reacted with a CO_2/H_2O gas mixture at 1000°F and 10 atm to convert the CaS to $CaCO_3$. A sample of the bed material was taken after each step in the cycle and analyzed for sulfur and sulfide content.

The effluent gas stream was analyzed for H_2S concentration, using the quadrupole mass spectrometer. A plot of H_2S concentration versus reaction time in the six cycles is presented in Figure 15.

The results indicated that the conversion of $CaSO_4$ to CaS in the reduction step was ineffective. Only in cycles 1 and 4 was the conversion to CaS greater than 50 percent. A possible cause could be the interaction of $CaSO_4$ and CaS to form nonporous surfaces; the formation of easily sinterable cakes has been reported when these materials are present.

The data for the regeneration step (presented in Figure 15) showed that the peak concentrations of H_2S in the effluent gas decreased from 13 volume percent (dry basis) for cycle 1 to 0.5 volume percent (dry basis) for cycles 5 and 6. The percent calcium sulfide converted to $CaCO_3$ decreased to a very low indeterminate value after several cycles. It appears from these data that a layer of material of low permeability is built up on or within pores of the additive particles, inhibiting the removal of sulfur. The high sulfur loading of the bed particles in these experiments may be a factor in the poor conversion.

In continuing work, it is planned to investigate the use of a high reduction temperature (e.g., 1800°F) to promote more complete reduction and to remove part of the sulfur as SO_2. The remaining sulfur would then be removed as H_2S by reaction with CO_2-H_2O. The two gas streams could be combined for conversion to elemental sulfur.

PRESSURIZED COMBUSTION AND REGENERATION—PILOT PLANT DESCRIPTION

Equipment has been installed for combusting coal at pressures up to 10 atm and for continuously regenerating sulfated lime for reuse. A simplified equipment schematic is shown in Figure 16. The regenerator and the fluidized-bed combustor have a common off-gas system (cyclones, filters, gas-sampling equipment, pressure let-down valve, and scrubber) and will not be operated simultaneously. Either the combustor or regenerator will be disconnected from the off-gas line and flanged off when the other unit is in operation.

The combustion unit consists of a 6-in. schedule 40 pipe (Type 316 SS) approximately 11 ft long, with an outer shell consisting of 12-in. schedule 10 pipe (Type 304 SS) over nearly the entire length. A bellows expansion joint is incorporated into the outer shell to accommodate the differential thermal expansion of the inner and outer vessels.

The unit is of a balanced pressure design; i.e., the annular chamber between the two pipes is maintained under pressure so that a differential pressure does not exist across the hot inner pipe wall. The balancing pressure for the shell is supplied by a bank of nitrogen cylinders.

A bubble-cap-type gas distributor is flanged to the bottom end of the inner vessel; thermocouples, solids feed lines, and solids take-off lines extend through the gas distributor. The outer wall of the 6-in. pipe is wrapped alternately with sixteen 3000-W tubular resistance heaters and 3/8-in. OD cooling coils that are spray-metal-bonded. Internal cooling coils of 3/8-in. pipe extend down into the interior of the 6-in. vessel from the flanged top to provide additional heat transfer area. Water flow to the cooling coils is regulated using flow indicators and is adjusted on the basis of the temperatures of the fluidized bed and reactor wall.

Both the annular pressure chamber and the reactor itself are equipped with rupture disc assemblies and pressure relief valves vented to the room ventilation exhaust ducts.

The regenerator has a -in. ID surrounded by 2-1/2 inches of Plibrico castable refractory and encased in an 8-in. schedule 40 pipe (316 SS). This entire assembly is enclosed by a pressure shell made of 12-in. schedule 20 carbon steel pipe. Differential thermal expansion between the inner and outer pipes is accommodated by the use of packing glands on the lines entering the bottom flange of the unit. The unit is of a balanced pressure design; i.e., the annular chamber between the two pipes is maintained under pressure so that a large differential pressure does not exist across the hot inner pipe wall. The balancing gas is nitrogen. Since the annular space is not gas tight with respect to the regenerator inner vessel, the pressure in the annular space will be maintained slightly higher than the regenerator pressure to prevent process gases from entering the annulus. A pressure alarm gauge will monitor the pressure in the annular space and will be set to warn of both high and low pressure.

A bubble-cap-type gas distributor is connected to the bottom of the inner vessel via a slip fit and held in place with retaining screws. Thermocouples, solids feed lines, and solids take-off lines pass through the gas distributor and then through packing glands on the bottom flange of the outer pressure vessel. The wall of the inner vessel is wrapped alternately with 3000-W tubular resistance heaters and 3/8-in. OD tubing coils. Both the annular chamber and the regenerator itself are equipped with rupture disc assemblies and pressure relief valves vented to the room ventilation exhaust ducts.

The primary filter cartridges are suitable for temperatures up to 350°F (epoxy-impregnated cellulose-base material with glass fiber substrate). The secondary filter cartridges are Rigimesh (woven metal wire).

The gas preheater is of a balanced pressure design and was designed in accordance with ASME code requirements . The design rating of the unit is 150 psig at 1500°F. Air (or gas) passes through an annulus, reverses direction, and passes through a heated section.

The feeders are of the rotary pocket type equipped with hoppers.

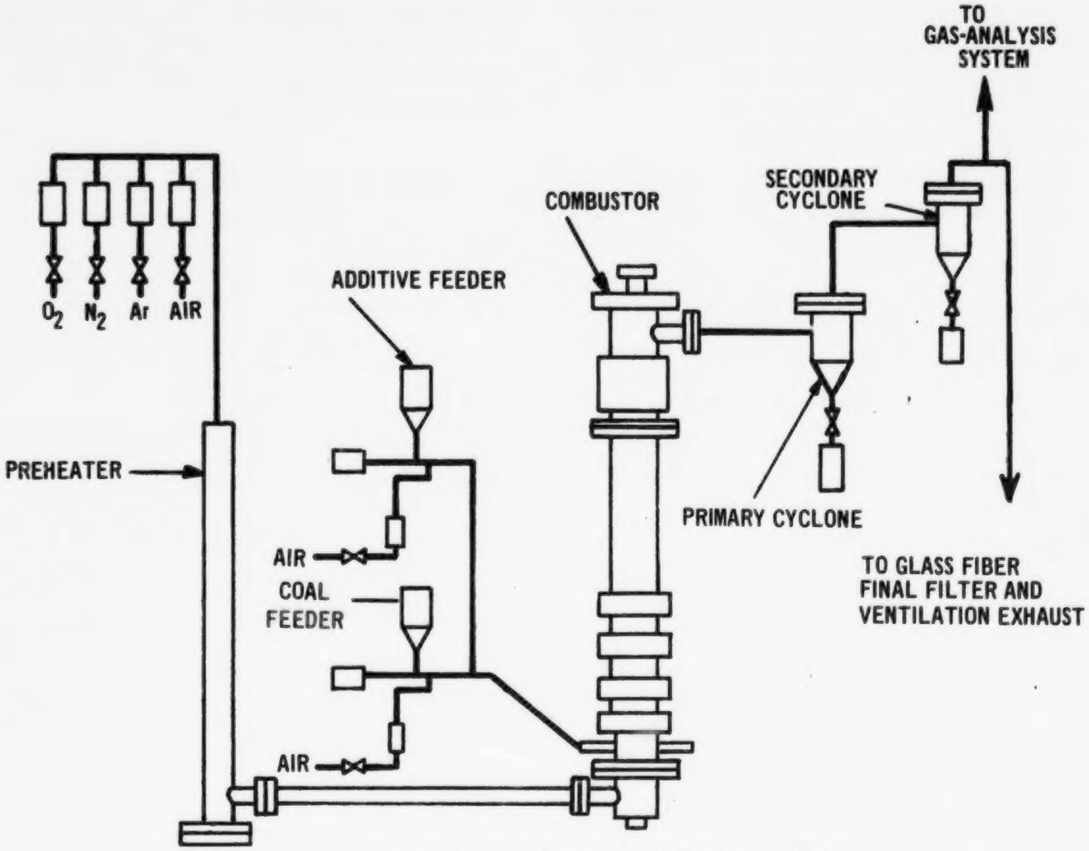

Figure 1. Bench-scale combustor system.

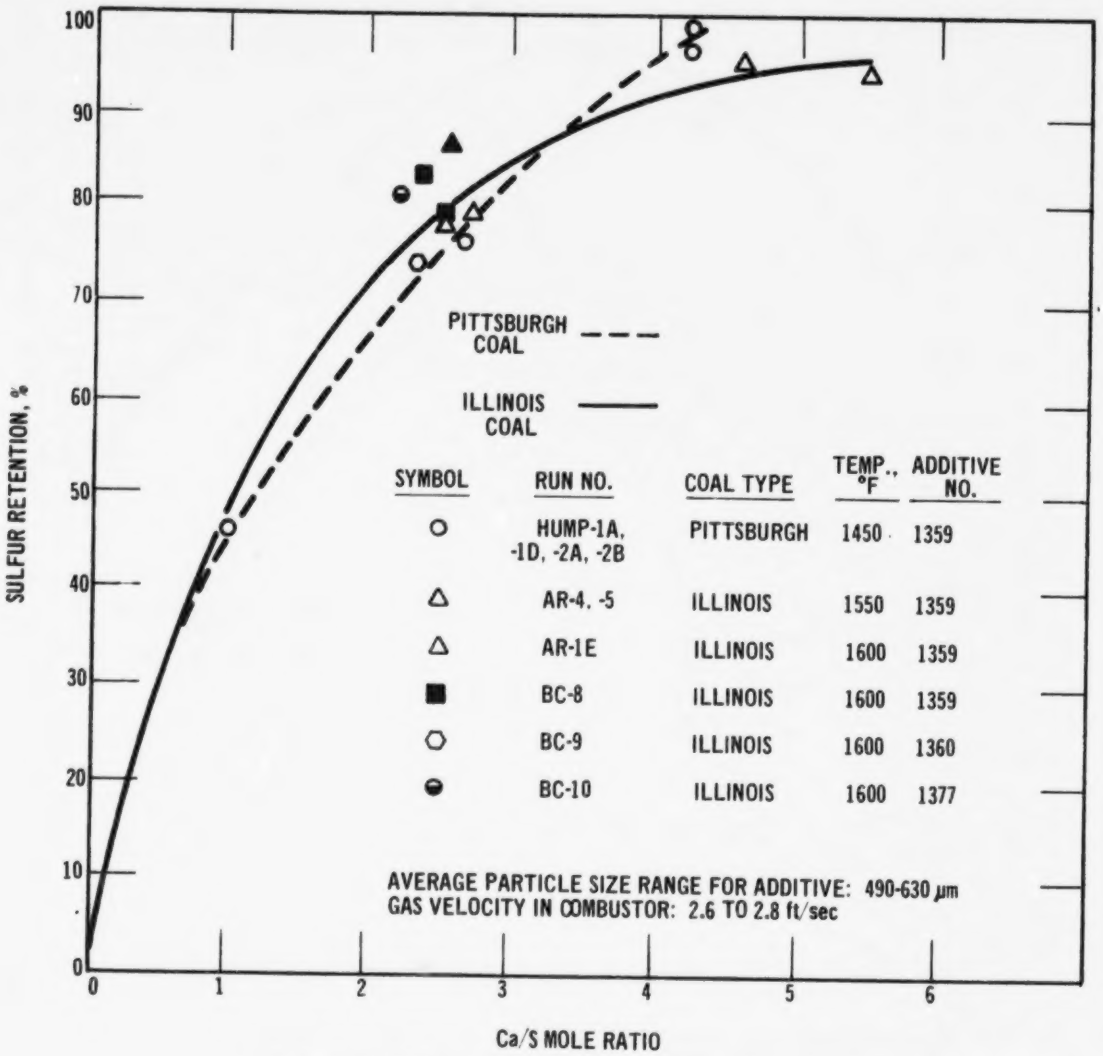

Figure 2. Effect of Ca/S mole ratio on sulfur retention.

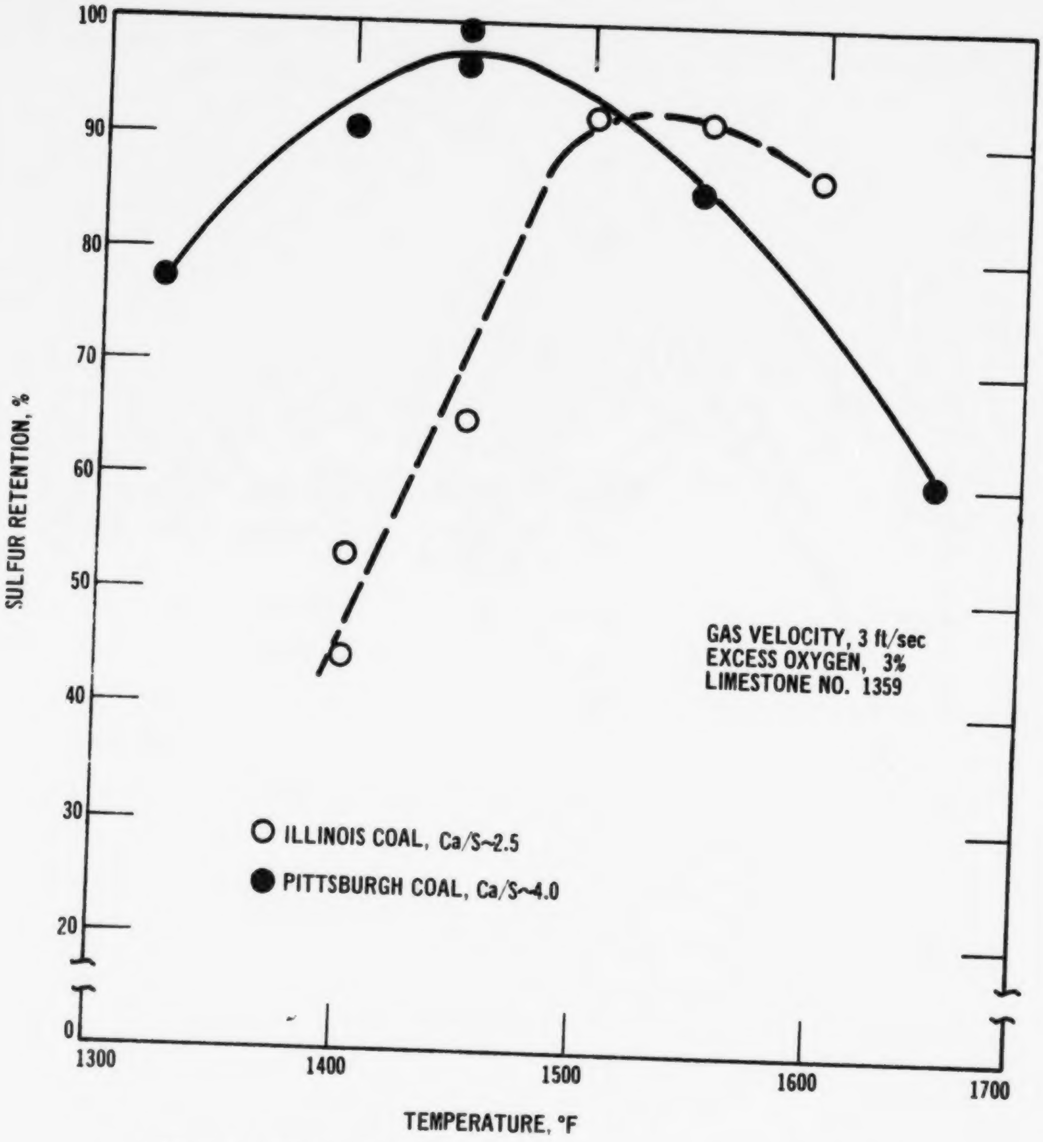

Figure 3. Effect of fluidized-bed temperature on sulfur retention.

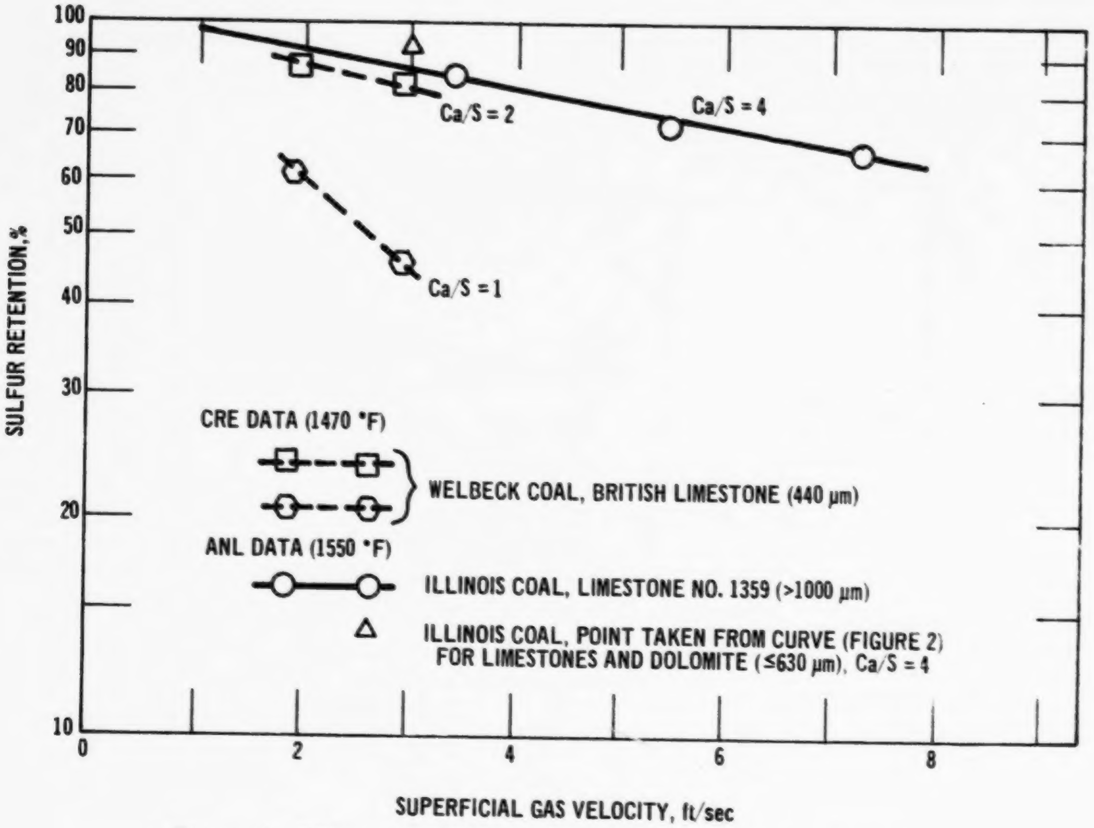

Figure 4. Effect of superficial gas velocity on sulfur retention.

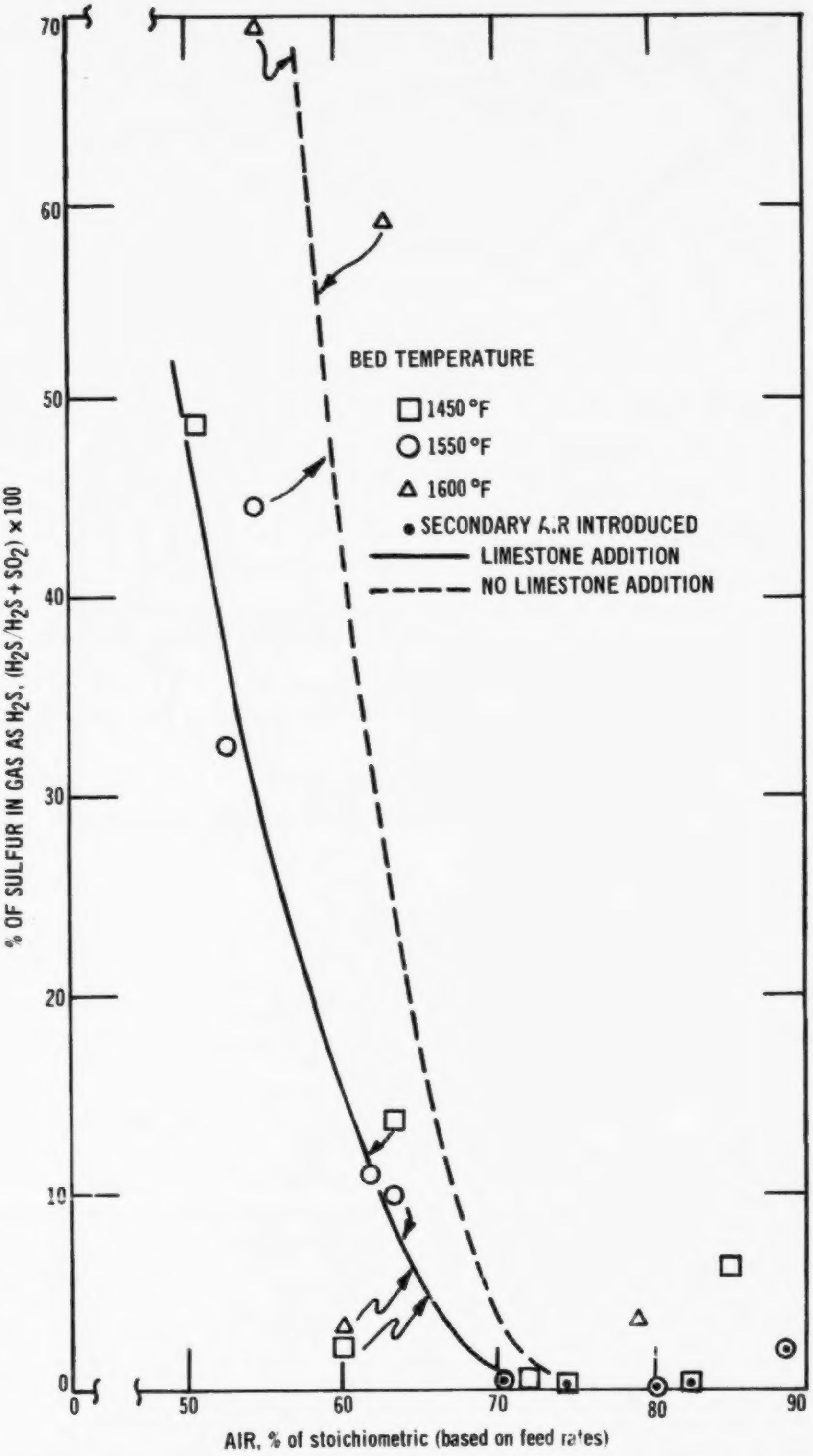

Figure 5. Effect of air feed rate on percent of sulfur in off-gas as H_2S.

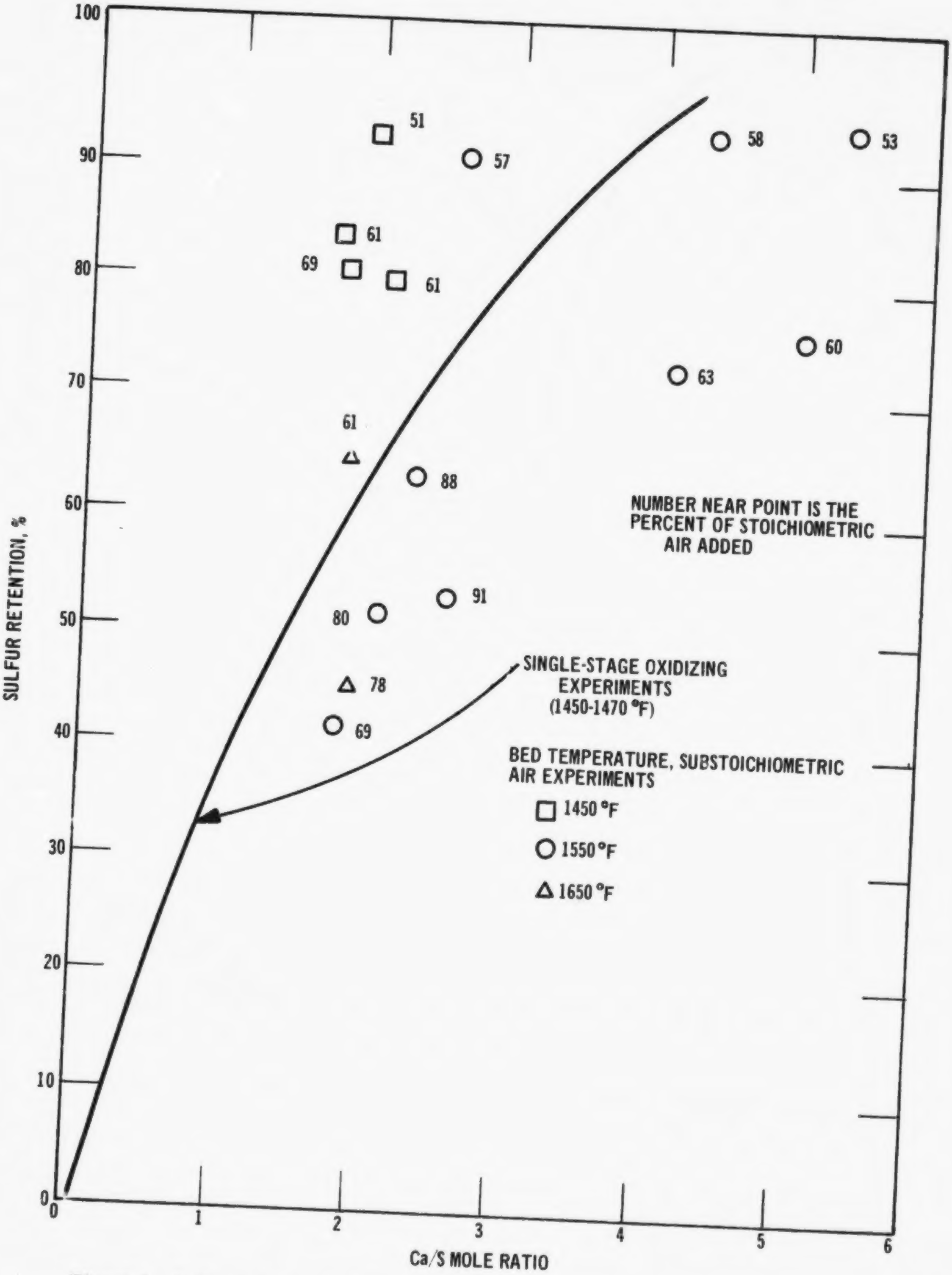

Figure 6. Sulfur retention in oxygen-excess and oxygen-deficient experiments.

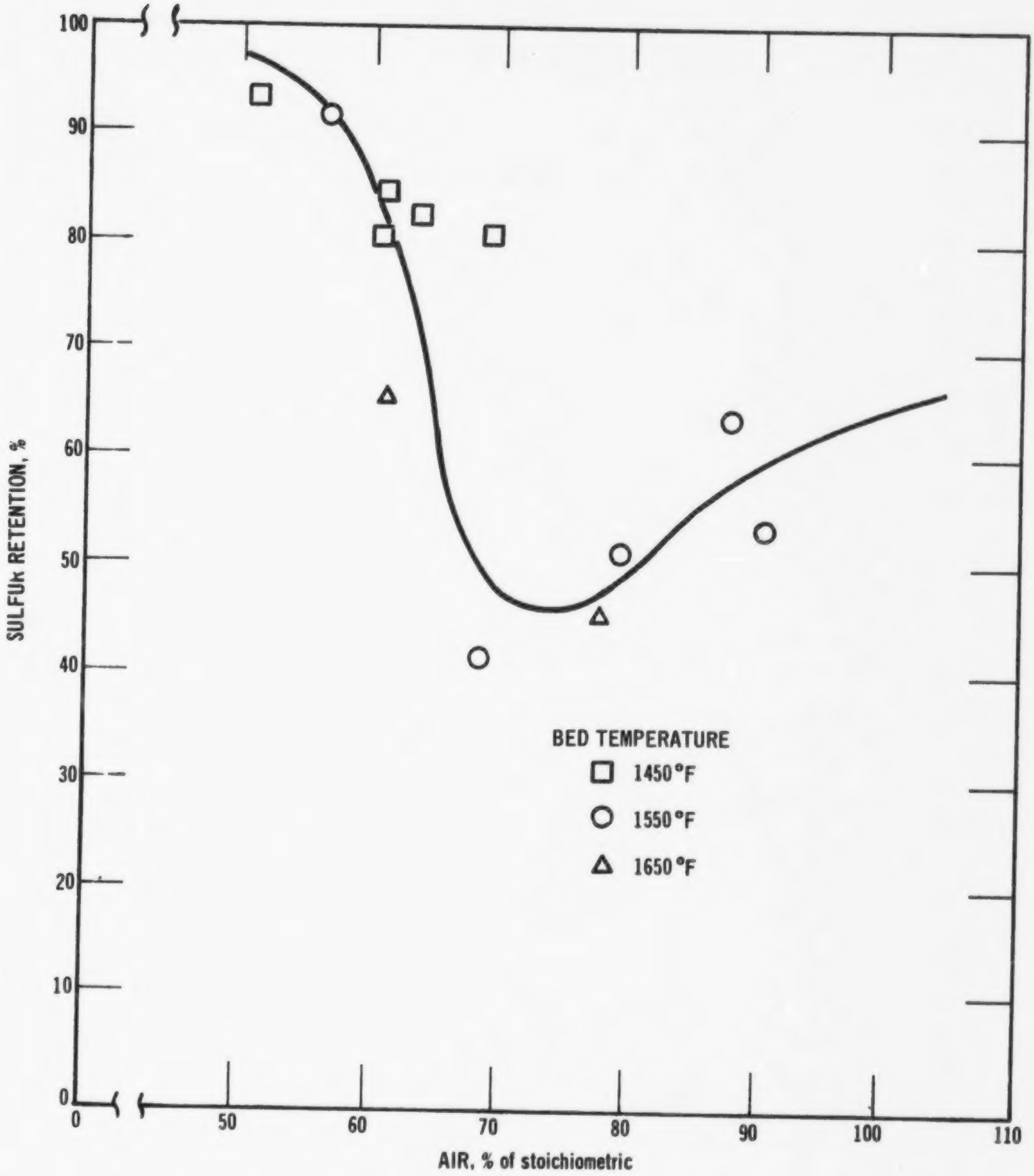

Figure 7. Effect on sulfur retention of air-feed rate to first stage.

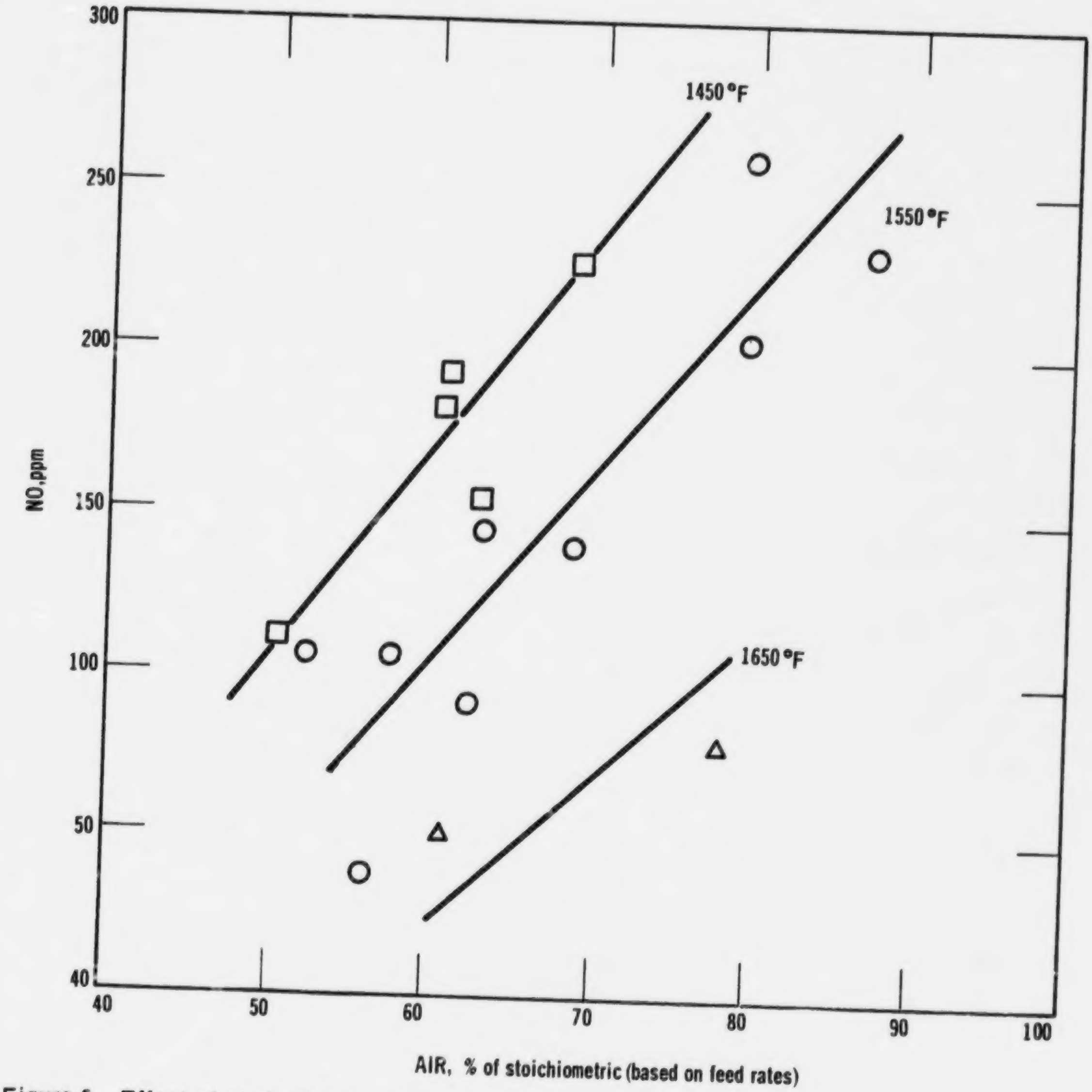

Figure 8. Effect of air feed rate and fluidized-bed temperature on NO concentration in off-gas from the first stage during combustion of coal.

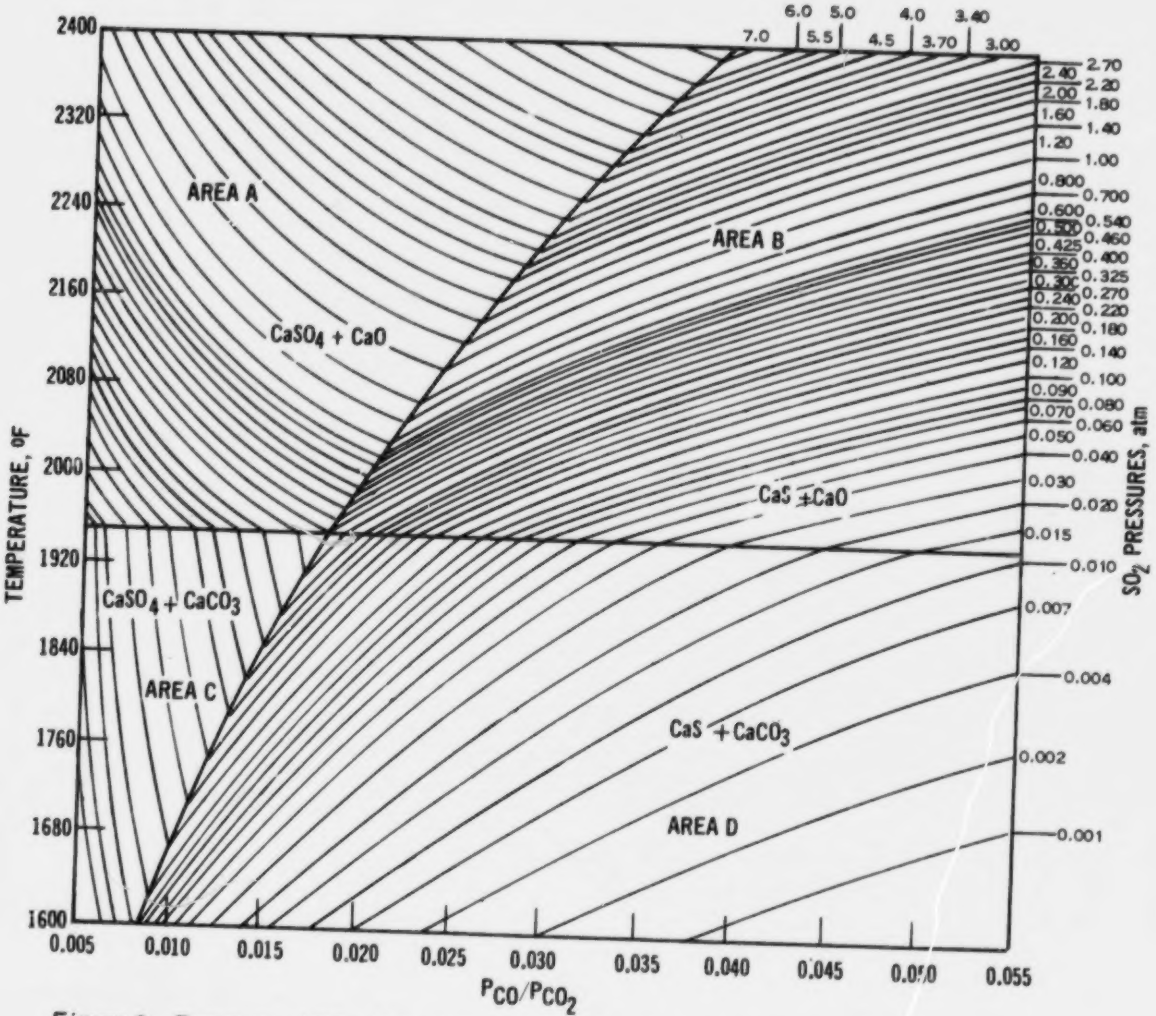

Figure 9. Pressure of SO_2 in equilibrium with CO/CO_2 mixtures as a function of temperature (10 atm total pressure).

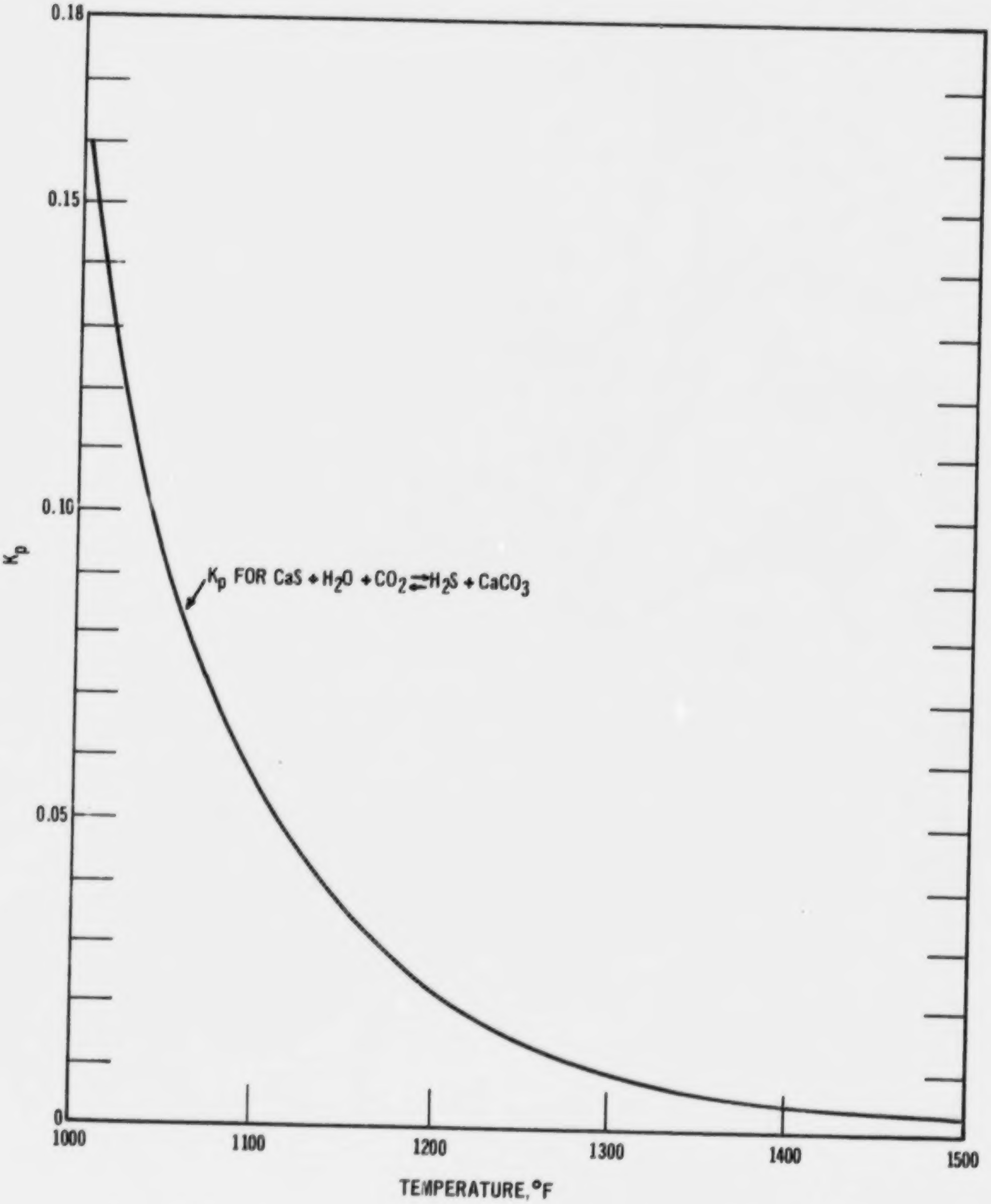

Figure 10. Equilibrium constant as function of temperature, $CO_2 + H_2O + CaS \rightarrow CaCO_3 + H_2S$.

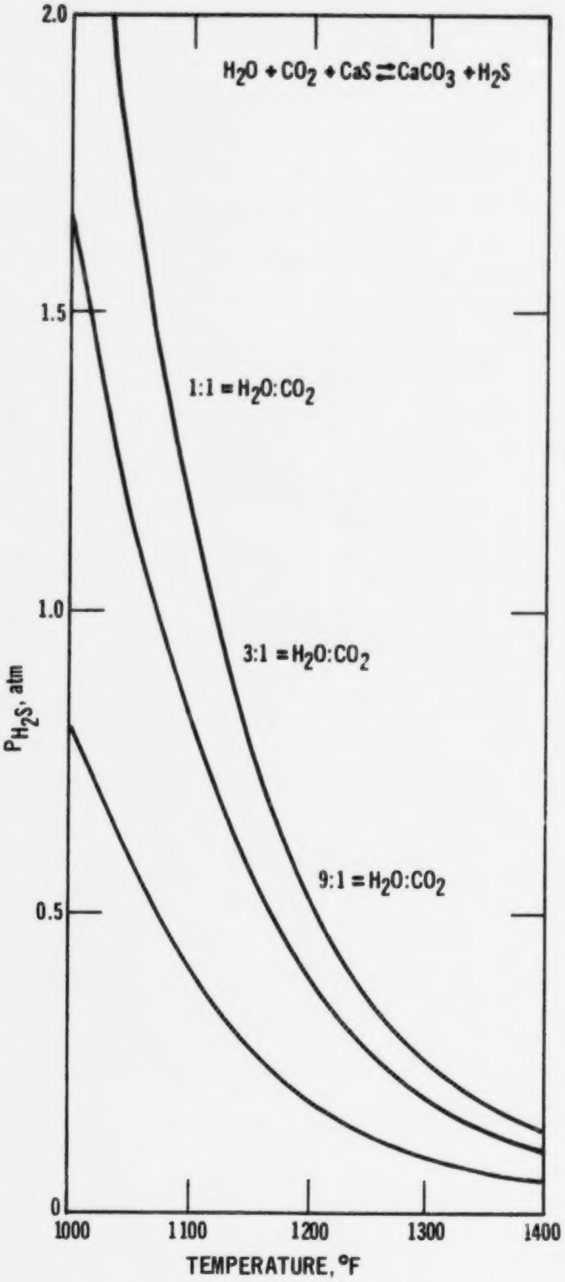

Figure 11. Pressure of H_2S in undried gas stream as function of temperature at 10-atm P_{total}.

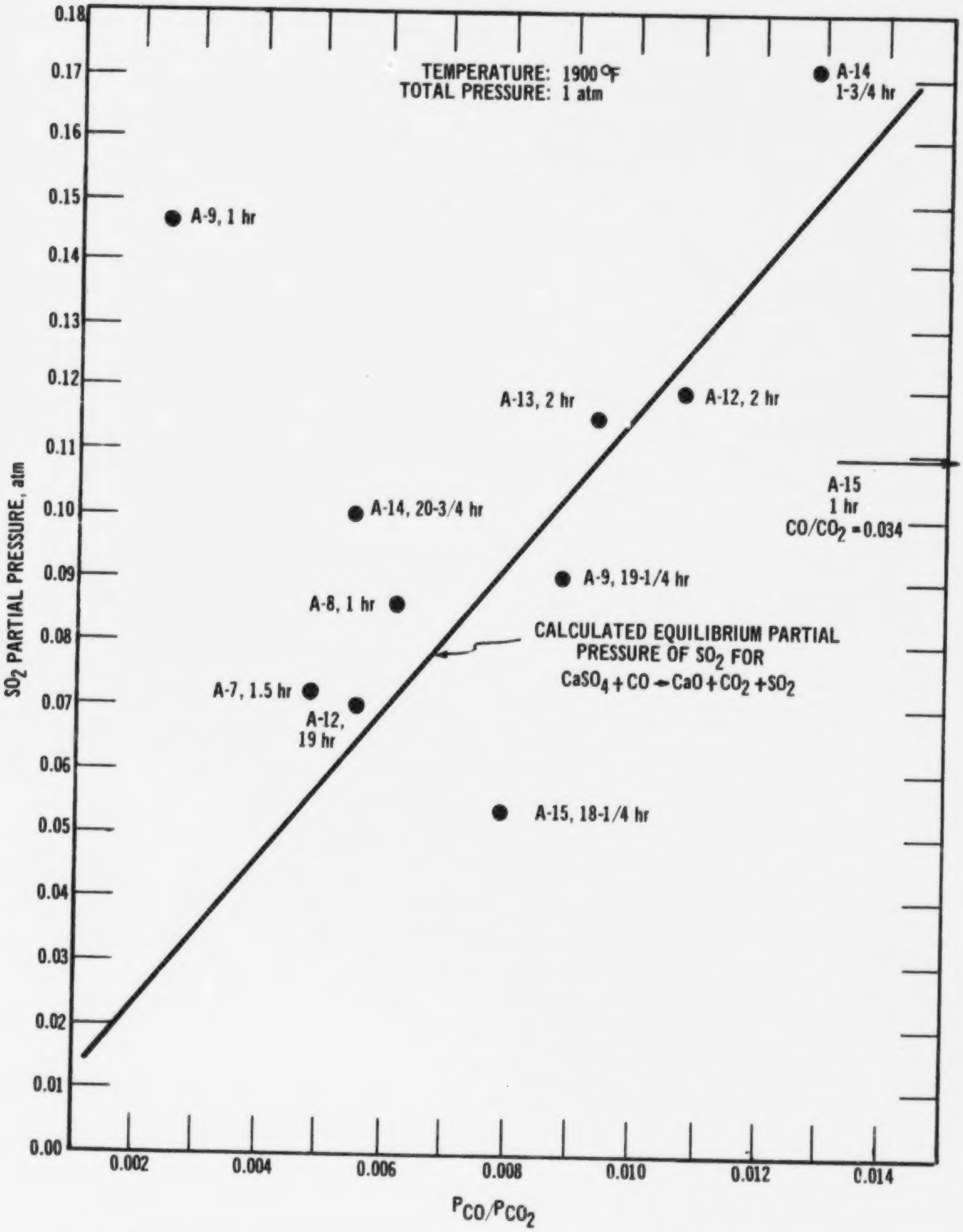

Figure 12. Partial pressures of SO_2 over a range of P_{CO}/P_{CO_2} ratios.

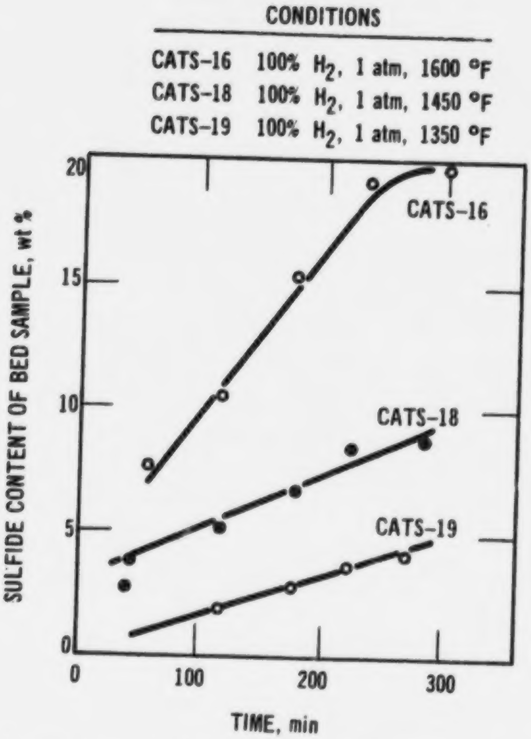

Figure 13. Sulfide content of bed during reduction of partially sulfated dolomite with hydrogen at various temperatures.

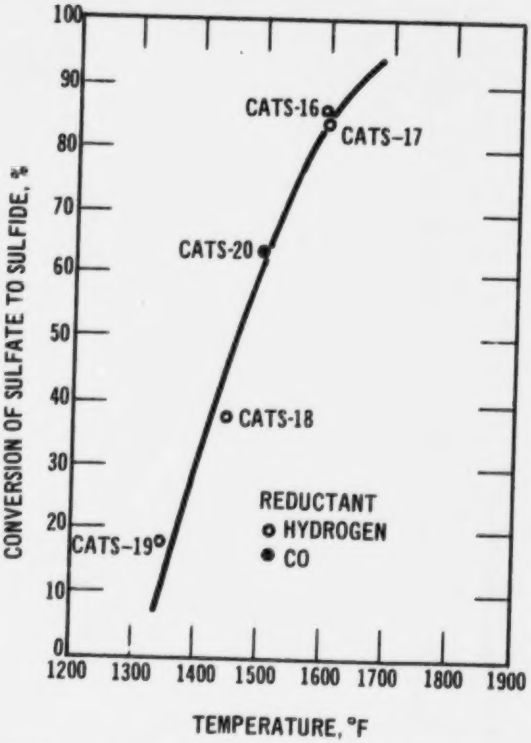

Figure 14. Effect of temperature on reduction of $CaSO_4$ (dolomite), 4.5 hours of reduction time.

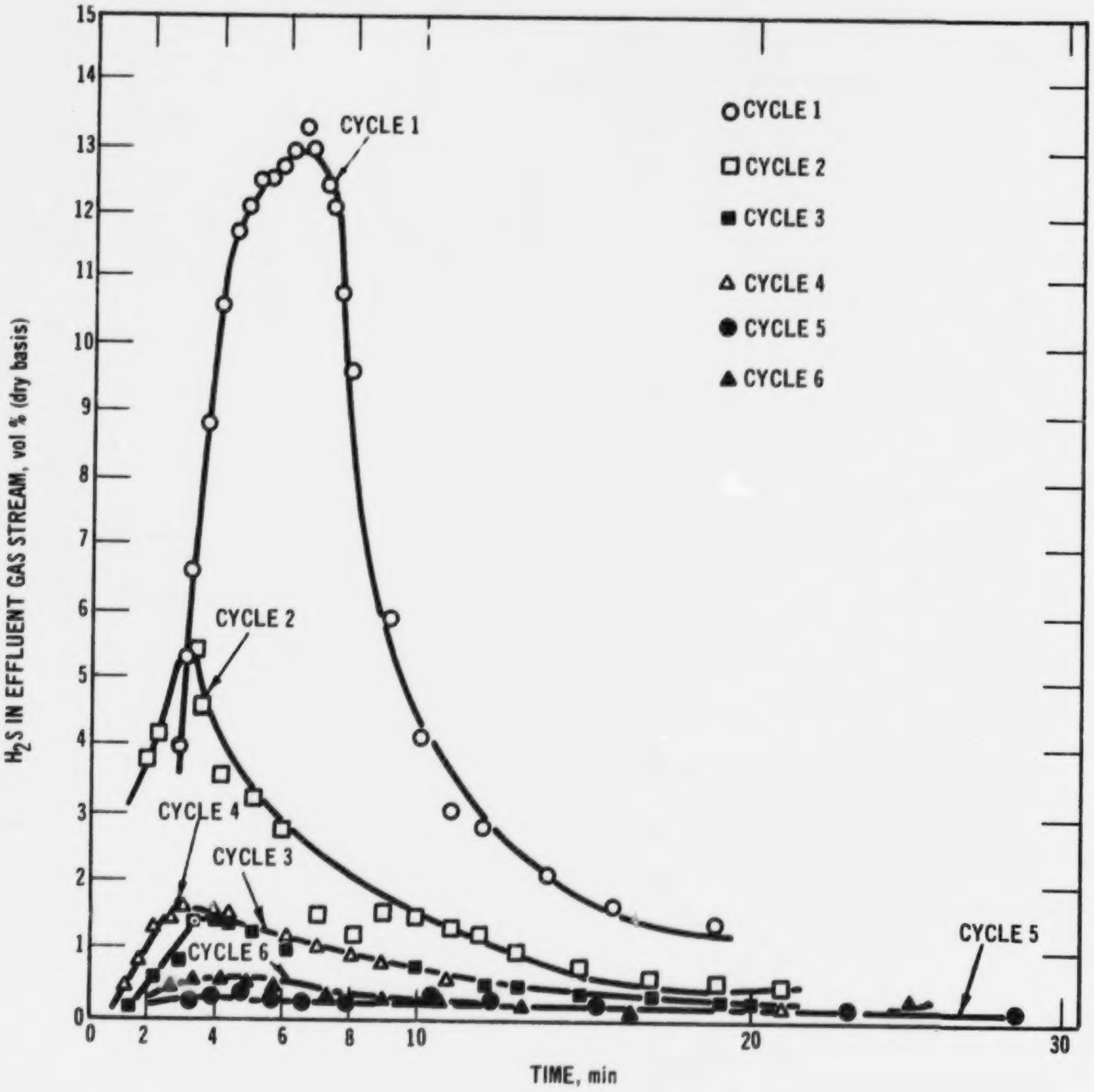

Figure 15. H_2S concentration in effluent gas stream in regeneration step.

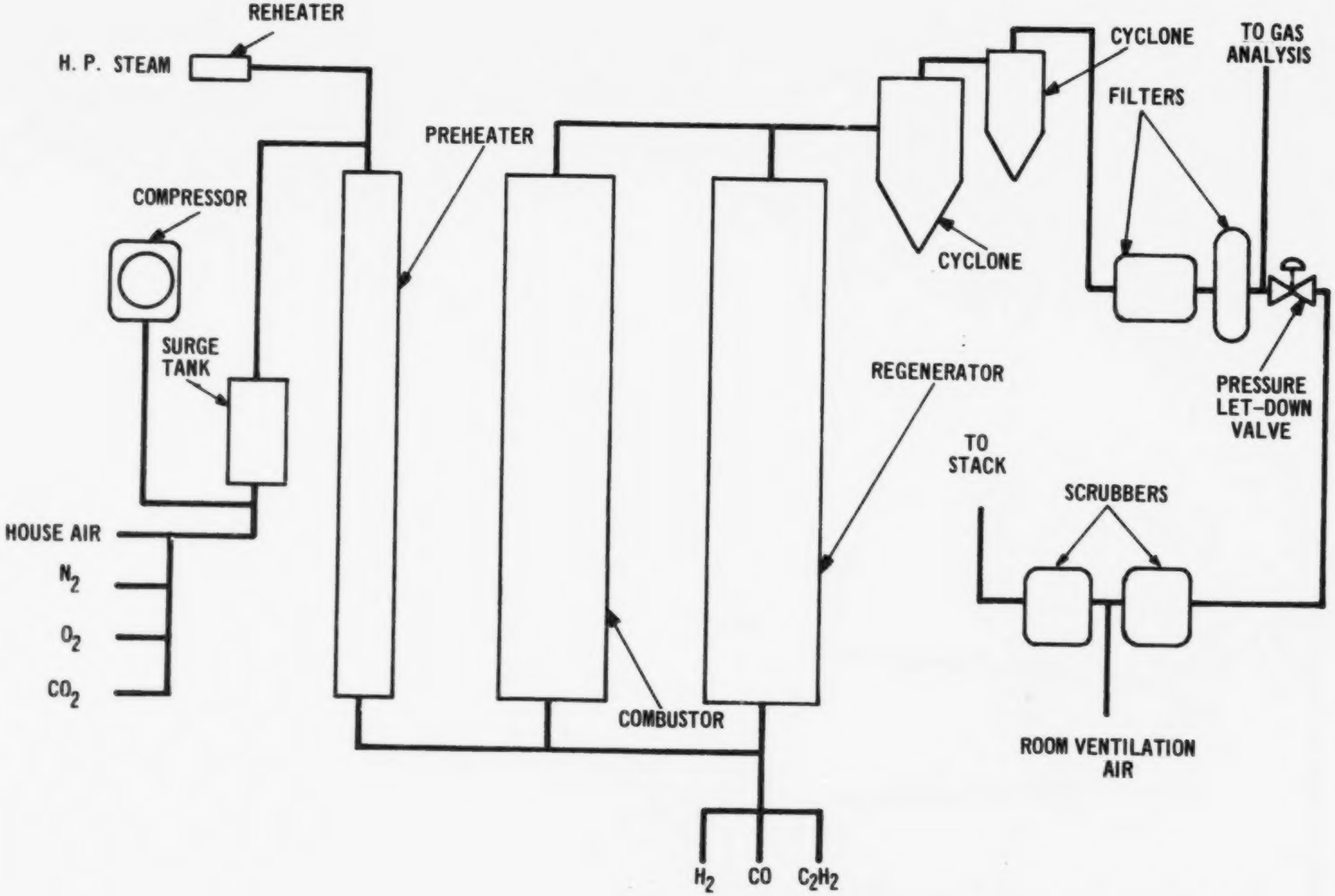

Figure 16. Simplified schematic of combustion-regeneration equipment.

2. A REGENERATIVE LIMESTONE PROCESS FOR FLUIDIZED-BED COAL COMBUSTION AND DESULFURIZATION

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ABSTRACT

The factors influencing NO_x emissions from a fluidized limestone bed coal combustor were studied. NO_x emissions were decreased by a decrease in temperature and a decrease in excess air. Sulfated lime depressed NO_x emissions compared to an inert alundum bed. An apparent cause of these effects is reduction of NO_x by CO. The NO/CO reaction was then studied further in fixed bed units. $CaSO_4$ catalyzed the reaction slightly compared to alundum in a dry system. CaO promoted the reaction significantly, giving over 90 percent conversion in the absence of CO_2. CO_2 was found to inhibit the rate, possibly due to a kinetic limitation caused by the presence of the CO_2. Study of the reaction of SO_2 and NO indicated that the reaction is catalyzed by partially sulfated lime, but not by $CaSO_4$ or alundum. Temperature was found to have a negative effect on the reaction, apparently due to the thermal instability of an intermediate, $CaSO_3$. Two-stage combustion of coal was studied to promote the CO/NO reaction and reduce NO emissions further. NO emissions were reduced by two-stage combustion, and the reduction was enhanced by operating the first stage at lower air levels.

Regeneration of $CaSO_4$ to CaO and SO_2 by CO and H_2 was studied at pressures up to 9.5 atm. SO_2 levels in the off-gas as high as 7.5 percent were measured at pressures up to 6 atm. The maximum concentration measured to date at 9.5 atm is 2.2 percent. The measured levels are 50-60 percent of the levels calculated from equilibrium considerations.

INTRODUCTION

Esso Research and Engineering Company is conducting an experimental program for the Environmental Protection Agency under contract CPA 70-19 to develop a regenerative limestone process for fluidized-bed coal combustion and desulfurization. This is a part of EPA's overall program to examine fluidized-bed combustion as a possible new power generation technique. The potential of fluidized-bed combustors for air pollution control is good because the intimate gas-solid contacting in a fluidized bed promotes high SO_2 removal efficiency on suitable materials such as limestone or dolomite.

A schematic diagram of the process is shown in Figure 1. In the combustor, the sulfur in the coal is burned to SO_2 which then reacts with the lime to form $CaSO_4$. The system being studied by Esso involves transferring the partially sulfated lime from the combustor to a separate regeneration vessel where the sulfated lime is regenerated according to the reaction

$$CaSO_4 + \underset{H_2}{CO} \rightarrow CaO + SO_2 + \underset{H_2O.}{CO_2} \qquad (1)$$

Previous Studies

Various laboratories including Esso Research have studied fluidized-bed coal combustion over the past few years. The results of the studies are summarized in Table 1 and have shown that coal can be burned efficiently with over 90 percent removal of SO_2 and with reduced NO_x emissions. Regeneration of sulfated limestone has been studied using a number of regeneration methods. The method studied at Esso Research consisting of the one step reduction of $CaSO_4$ to CaO and SO_2 gives 6-10 percent SO_2 in the product gas when carried out at 1 atm and about 2000°F. The recycled lime was also shown to maintain a reasonably high level of activity after seven combustion/regeneration cycles.

Table 1. **PREVIOUS FBC FINDINGS**

Coal combustion efficiency high

Over 90% removal of SO_2

NO_x emissions reduced

Sulfated lime can be regenerated

$$CaSO_4 + CO \rightarrow CaO + SO_2 + CO_2$$

Activity maintenance of recycled lime satisfactory after 7 cycles

Pressurized FBC system more attractive

Economic studies were carried out by Westinghouse Research Laboratories under contract to EPA.[3] These studies indicated that operation of the combustor and regenerator at higher pressures, approximately 10 atm, would be significantly more economical than atmospheric pressure operation. As a result, the current studies are being made at higher pressures.

Objectives

Objectives of Esso Research's current experimental program are summarized in Table 2 and consist of (1) investigating the factors influencing the reduction of NO_x emissions in fluidized-bed combustion, and (2) studying the regeneration of sulfated lime at pressures up to 10 atm. The latter objective has required construction of higher pressure experimental equipment.

Table 2. **OBJECTIVES OF CURRENT EXPERIMENTAL WORK**

1. Investigate factors influencing reduction of NO_x emissions
   - Effect of temperature, excess air, bed materials
   - Reaction of NO with CO and SO_2
2. Study regeneration of sulfated lime
   - SO_2 levels attainable at higher pressures
   - Kinetics of regeneration and stone activity maintenance

EXPERIMENTAL EQUIPMENT

A number of experimental units were used in the current program. A flow diagram of the atmospheric pressure fluidized-bed combustion is shown in Figure 2. The reactor is a 3-in.-ID Incoloy tube. Four continuous flue gas analyzers are used including IR SO_2 and CO analyzers and polarographic NO_x and O_2 analyzers. A high pressure regeneration unit capable of operating up to 10 atm was recently built and is shown in Figure 3. The reactor consists of a 3-in.-ID alumina tube contained in a 12-in. carbon steel vessel. The reactor is 15 feet long. The interior of the steel vessel is lined with 4-1/2 inches of castable refractory insulation. The regeneration feed gas is produced by combustion of propane. N_2 and CO_2 can also be added to the burner to adjust the composition of the regeneration gas. Most of the input heat is provided by combustion of propane, but additional heat input is provided through electrical heaters adjacent to the alumina tube and an air preheater. The unit is heated by burning the propane under excess air conditions. When the operating temperature has been reached, the air/fuel ratio is instantaneously changed to substoichiometric conditions and N_2/CO_2 flow is started, thereby assuring a rapid change from heat-up to operating conditions.

Two small fixed bed units were also used in these studies. Three different reactors were used: a 2-1/2-in. alumina tube operating at 1 atm, a 1-in. stainless steel tube operating at pressures up to 10 atm, and a specially constructed regenerator. The regenerator consisted of a 1-in. alumina tube contained in a 3-in. pipe with fiber insulation between the pipe and tube. This unit was capable of operating at pressures up to 9.5 atm at temperatures up to 2000°F. All fixed bed units were electrically heated.

EXPERIMENTAL RESULTS

Factors Affecting NO_x Emissions

It was determined previously that NO_x emissions measured at the low temperatures occurring in fluidized-bed combustion are formed by oxidation of nitrogen compounds in the coal. Oxidation of atmospheric N_2 occurs only at higher temperatures. In this study, the effects of temperature, excess air, and fluidized bed material on NO emissions were measured. The effect of temperature using a bed of $CaSO_4$ in the combustor is shown in Figure 4. As temperature decreased, NO emissions dropped rather sharply below 1500°F. The effect of excess air using a bed of $CaSO_4$ is shown in Figure 5. Actual NO emissions decreased as excess air (percent O_2) was increased. However, when the emissions were normalized to a constant gas volume (at 3 percent O_2), the NO emissions increased as the excess air increased. The NO formation rate was thus increased by the higher average oxygen concentration in the bed. The effect of bed material is shown in Figure 6. $CaSO_4$ gave lower emissions than alundum. With a CaO bed the emissions were high initially, but as the bed sulfated the emission level approached that of $CaSO_4$.

One consistent explanation for these results is the reaction of NO with CO. Carbon monoxide emissions are higher at the lower temperatures and at lower excess air conditions. The higher CO levels then give lower NO emissions. The effect of bed materials appears to be a catalytic effect.

Reactions of NO and CO

The reaction of CO and NO was studied further in fixed-bed units. The effects of bed material, temperature and feed gas composition were studied. In a dry system, $CaSO_4$ catalyzed the reaction slightly and showed a small effect of temperature, but alumina and an empty bed gave essentially no reaction. This is shown in Table 3. However, the addition of water enhanced the reaction and gave the same NO conversion regardless of the presence of the bed material. This is shown in Table 4. But when CaO was used as the bed material in a dry system, a very rapid reaction occurred which gave over 90 percent conversion of the limiting reactant as shown in Table 5. The reaction proceeded in 1:1 mole ratio of CO and NO suggesting the reaction

$$2\,CO + 2\,NO \rightarrow 2\,CO_2 + N_2 \quad (2)$$

Table 3. NO-CO REACTIONS—EFFECTS OF TEMPERATURE AND BED MATERIAL

Bed material	$CaSO_4$	$CaSO_4$	Alumina	None
Bed temperature, °F	1500	1700	1500	1500
Inlet gas composition				
NO, ppm	1990	2025	2010	2035
CO, ppm	4000	4000	4000	4000
NO conversion, %	4	6	1	0.5

Table 4. NO-CO REACTIONS—EFFECT OF WATER VAPOR

Bed material	$CaSO_4$		Alumina		None	
Inlet gas composition						
NO, ppm	1990		2010		2035	
CO, ppm	4000		4000		4000	
H_2O, %	0	7.2	0	7.2	0	7.2
NO conversion, %	4	12	1	13	0.5	15

Temperature, 1500°F

Carbon dioxide was then added to the feed and reduced the conversion significantly over both calcined limestone and calcined dolomite. This is shown in Table 6.

Table 5. NO-CO REACTIONS—EFFECT OF CaO

Bed source	Lime # 1359		Dolomite # 1337	
Inlet gas composition				
NO, ppm	1400	1800	1400	1990
CO, ppm	940	1870	900	2080
Outlet gas composition				
NO, ppm	400	20	350	240
CO, ppm	10	160	20	100
Conversion, %	99	99	98	95

Temperature, 1600°F

Residence time, 0.3 sec

Table 6. NO-CO REACTIONS—EFFECT OF CO_2

Bed source	Lime # 1359		Dolomite # 1337	
Inlet gas composition				
NO, ppm	1400	860	1400	840
CO, ppm	940	990	900	980
CO_2, %	0	17	0	16
Conversion, %	99	26	98	19

Temperature, 1600°F

Residence time, 0.3 sec

The effect of pressure was studied and although increasing the pressure to 10 atm in the presence of CO_2 apparently increased the conversion, the increase was probably due to increased residence time. At equivalent residence times, increasing pressure appeared to decrease the conversion slightly. This is shown in Table 7. Changing the background gas from N_2 to argon appeared to increase the conversion very slightly, but the effect may not be significant. Oxygen was added to the feed and appeared to increase conversion slightly. This is shown in Table 8.

Table 7. NO-CO REACTIONS—EFFECT OF PRESSURE AND RESIDENCE TIME

Bed source	Lime # 1359			Dolomite # 1337	
Pressure, atm	1	10	10	1	10
Residence time, sec	0.3	3	0.3	0.3	3
Inlet gas composition					
NO, ppm	860	890	1150	840	840
CO, ppm	990	980	1240	980	980
CO_2, %	17	18	13	16	16
Conversion, %	26	82	16	19	88

Temperature, 1600°F

Table 8. NO-CO REACTIONS—EFFECT OF O_2

Bed source	Lime # 1359		Dolomite # 1337	
Inlet gas composition				
NO, ppm	1150	970	840	980
CO, ppm	1240	1060	980	1080
CO_2, %	13	16	16	15
O_2, %	0	2.4	0	2.3
Conversion, %	84	95	88	91

Temperature, 1600°F

Pressure, 10 atm

The most likely explanation for these effects is a kinetic limitation caused by the presence of the CO_2. Formation of $CaCO_3$ and inhibition caused by chemical reversibility were considered as possible explanations, but were ruled out after closer examination.

Reactions of NO and SO_2

Further studies of the reaction of NO and SO_2 were made in a fixed-bed reactor. The effects of bed material and temperature were studied. The effect of bed material is shown in Table 9. The results show that NO and SO_2 did not react in the vapor phase or over alundum or $CaSO_4$. However, a reaction did occur over partially sulfated lime and appeared to be dependent on SO_2 concentration. Further rate studies indicated a 0.5 order dependence on the NO concentration. Temperature had a negative effect on the rate, decreasing the rate with increasing temperature, as shown in Figure 7. A proposed mechanism for the reaction involves the reversible formation of $CaSO_3$ intermediate from CaO and SO_2. The sulfite then reacts

Table 9. NO-SO_2 REACTIONS—EFFECT OF BED MATERIAL AT 1600°F

Bed material	NO concentration, ppm		SO_2 concentration, ppm	
	Before SO_2 introduced	After SO_2 introduced	Inlet	Outlet
Gas phase	900	900	1290	1290
Partially sulfated limestone	840 830	440 180	780 1510	300 480
Alundum	820	820	1000	1000
$CaSO_4$	880	860	670	670

with NO to form N_2 and $CaSO_4$. However, it is known that the sulfite becomes unstable in the temperature range where the SO_2/NO reaction rate drops; this instability is the probable explanation for the negative temperature effect.

Two-Stage Combustion

The reactions of NO with CO suggest the possible lowering of NO emissions by operating a staged combustion system. Air would be injected at two points in the combustor giving an O_2 lean section at the bed inlet which should promote NO reduction because of the relatively high CO levels. The second step would then complete combustion. The fluid-bed combustor was modified to operate in a staged fashion by injecting second stage air 6 inches above the grid. The results of two runs are shown in Figure 8. As the ratio of the second stage air to the first stage air was increased, the NO emissions dropped. Nitric oxide emissions were lowered to 200 ppm. Although these conditions may not be feasible in commercial operation, the principle of stage combustion appears attractive.

Regeneration of Sulfated Limestone

Regeneration studies were carried out in fixed and fluidized beds using $CaSO_4$ at pressures up to 9.5 atm. The results are shown in Table 10.

Concentrations of SO_2 in the off-gas as high as 7.5 percent have been measured at pressures up to 6 atm. At 10 atm, the highest SO_2 concentration measured to date was a little over 2 percent. Comparisons were also made with SO_2 levels estimated from equilibrium calculations made by Argonne National Laboratory.[2] The equilibrium SO_2 partial pressure is determined by the temperature and the CO/CO_2 ratio in the gas in equilibrium with the solids. However, at each temperature, there is a CO/CO_2 ratio which gives the maximum attainable SO_2 partial pressure for the temperature in question. In the fixed-bed runs, the off-gases were not analyzed for CO and CO_2, and the measured SO_2 concentrations had to be compared to the maximum equilibrium SO_2 concentration. In the fluidized-bed runs, comparisons were made at the actual CO/CO_2 ratio measured, although these ratios were probably in error due to oxidation of CO in the exit lines from the reactor. The comparison of measured and calculated SO_2 levels is given in Table 10. The SO_2 levels measured in the fixed-bed unit were less than 50 percent of the maximum attainable at the temperatures of the runs. The results from the fluidized bed were closer to the equilibrium concentrations calculated for the CO/CO_2 ratio measured for each run. In general, the measured SO_2 concentrations were 50-60 percent of the equilibrium levels;

Table 10. REGENERATION OF SULFATED LIMESTONE, $CaSO_4$ AS BED MATERIAL

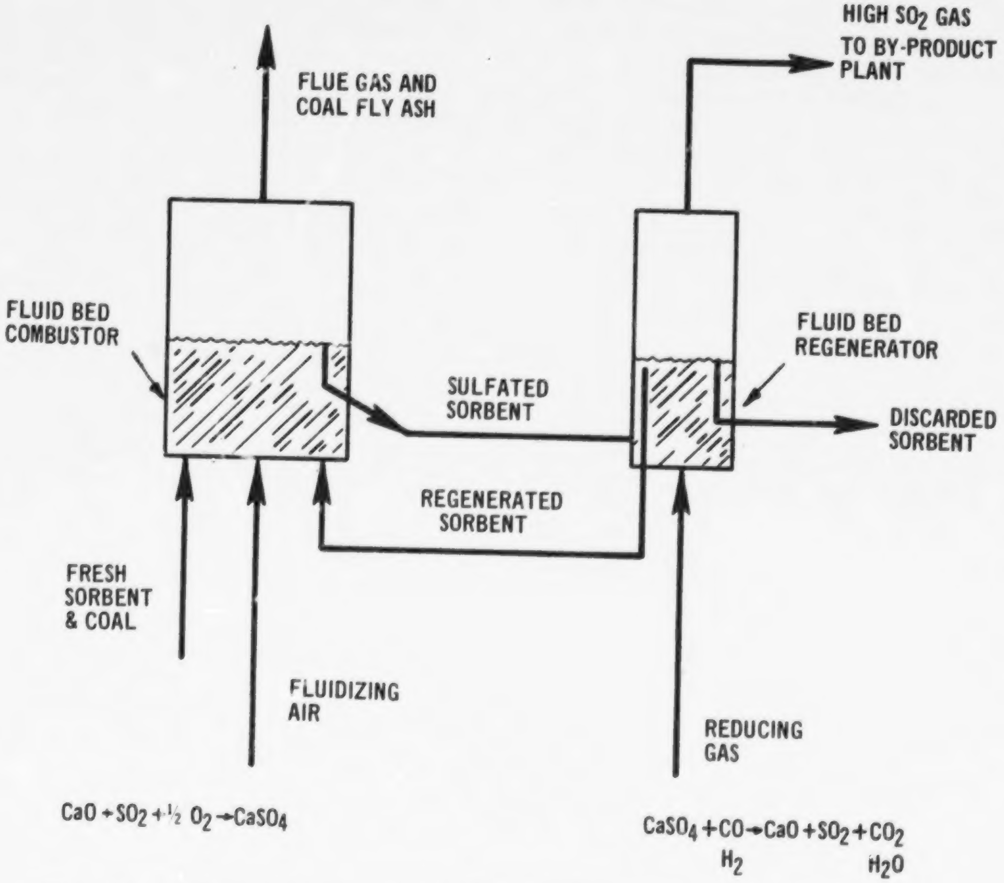
Unit	Pressure, atm	Temperature, °F	SO_2 concentration,% Measured	Calculated	SO_2 ratio measured/calculated
Fixed	3	2000	7.2	15.0	0.48
Fixed	9.5	2000	2.2	4.8	0.46
Fluidized	3.2	1990	5.2	9.8	0.53
Fluidized	6.2	2100	7.5	11.3	0.66
Fluidized	6.0	1950	2.0	3.7	0.54
Fluidized	6.0	1870	1.8	1.8	1.0

the last run met the calculated equilibrium concentrations. Further work is planned in the fluidized-bed regeneration unit to determine the SO_2 levels attainable at pressures up to 10 atm as a function of temperature, regeneration gas composition and flow rate, particle size, and sulfated lime source. A new pressurized combustor unit is being built which will be used with the regenerator to measure cyclic activity maintenance of various stones.

BIBLIOGRAPHY

1. Archer, D. H., D. L. Keairns, J. R. Hamm, R. A. Newby, W. C. Yang, L. M. Handman, and L. Elikan. Evaluation of the Fluidized-Bed Combustion Process. Westinghouse Research Laboratories, Pittsburgh, Pa. Prepared for the Environmental Protection Agency, Research Triangle Park, N. C. under Contract Number CPA 70-9. November 1971.

2. Jonke, A.A., G.J. Vogel, J. Ackerman, M. Haas, J. Riha, C.B. Schoffstoll, J. Hepperly, R. Green, and E.L. Carls. Reduction of Atmospheric Pollution by the Application of Fluidized-Bed Combustion. Argonne National Laboratory, Argonne, Ill. Prepared for the Environmental Protection Agency, Research Triangle Park, N.C. under agreement EPA-IAG-0020. Monthly Progress Report Number 38, December 1971.



FLUE GAS AND COAL FLY ASH
HIGH SO_2 GAS TO BY-PRODUCT PLANT
FLUID BED COMBUSTOR
FLUID BED REGENERATOR
SULFATED SORBENT
DISCARDED SORBENT
REGENERATED SORBENT
FRESH SORBENT & COAL
FLUIDIZING AIR
REDUCING GAS
$CaO + SO_2 + \frac{1}{2} O_2 \rightarrow CaSO_4$
$CaSO_4 + CO \rightarrow CaO + SO_2 + CO_2$
H_2
H_2O

Figure 1. Esso proposed fluidized-bed combustion-lime regeneration system.

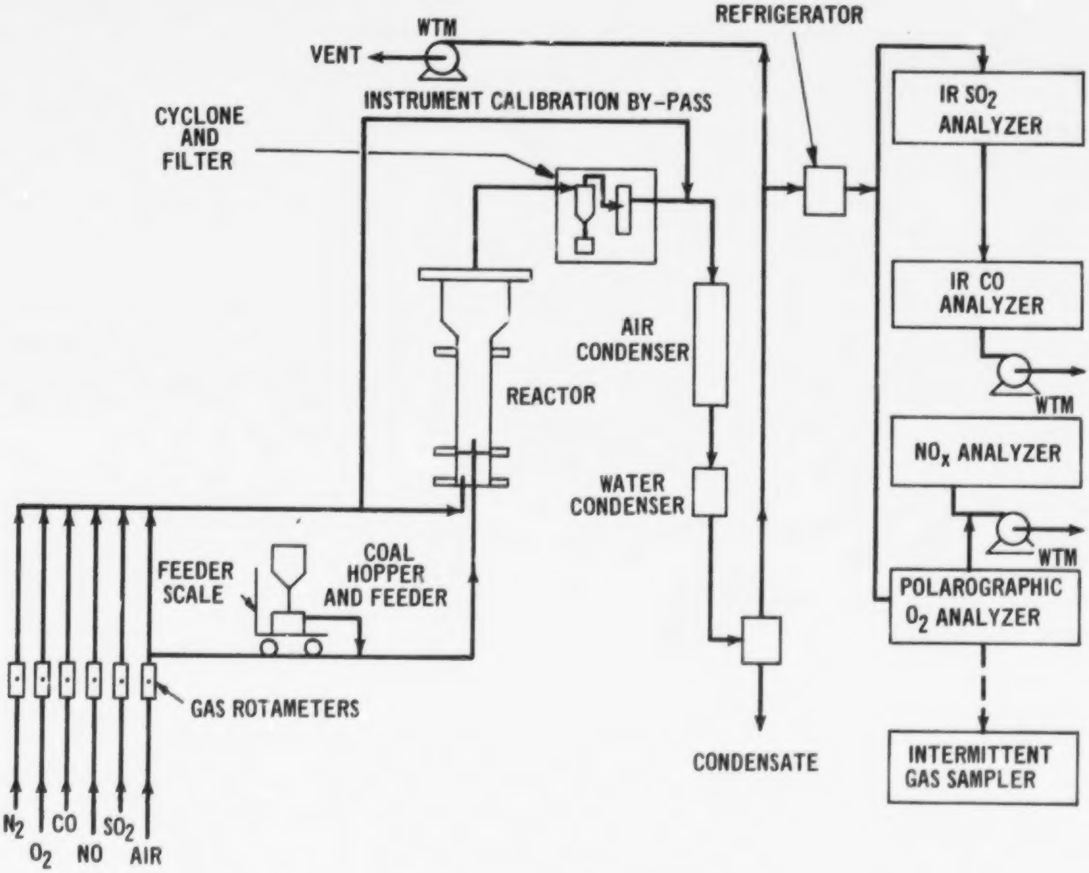


Figure 2. Esso fluidized-bed combustion unit.

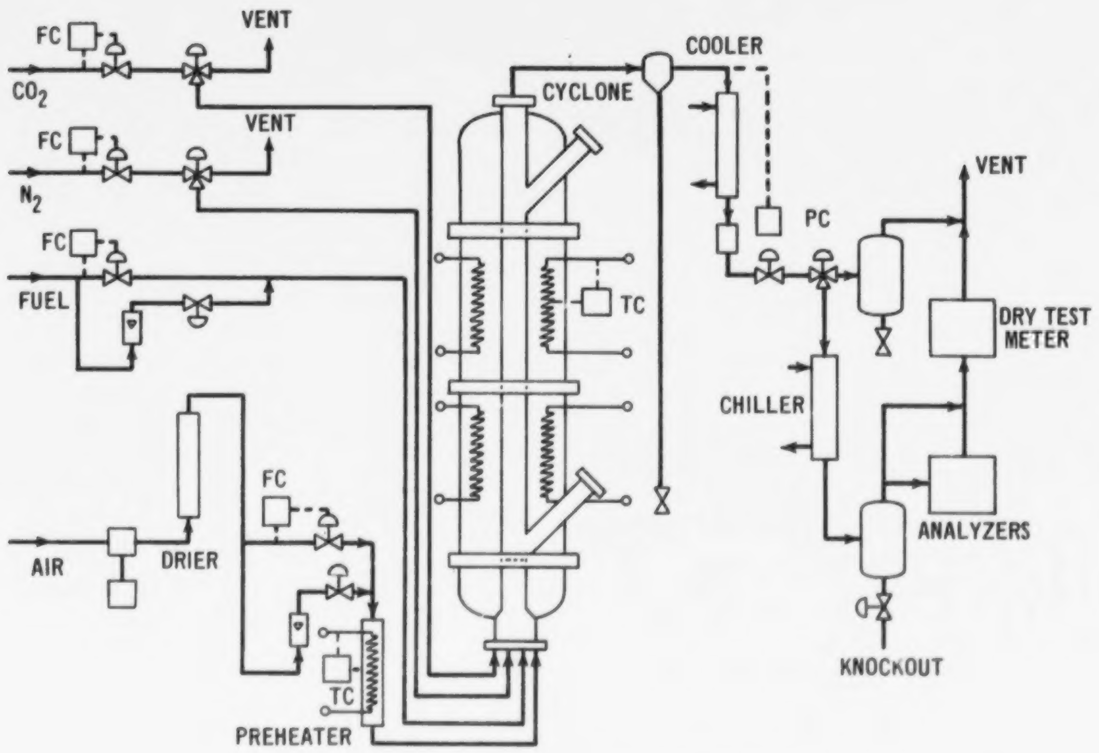


Figure 3. Fluidized-bed regeneration unit.

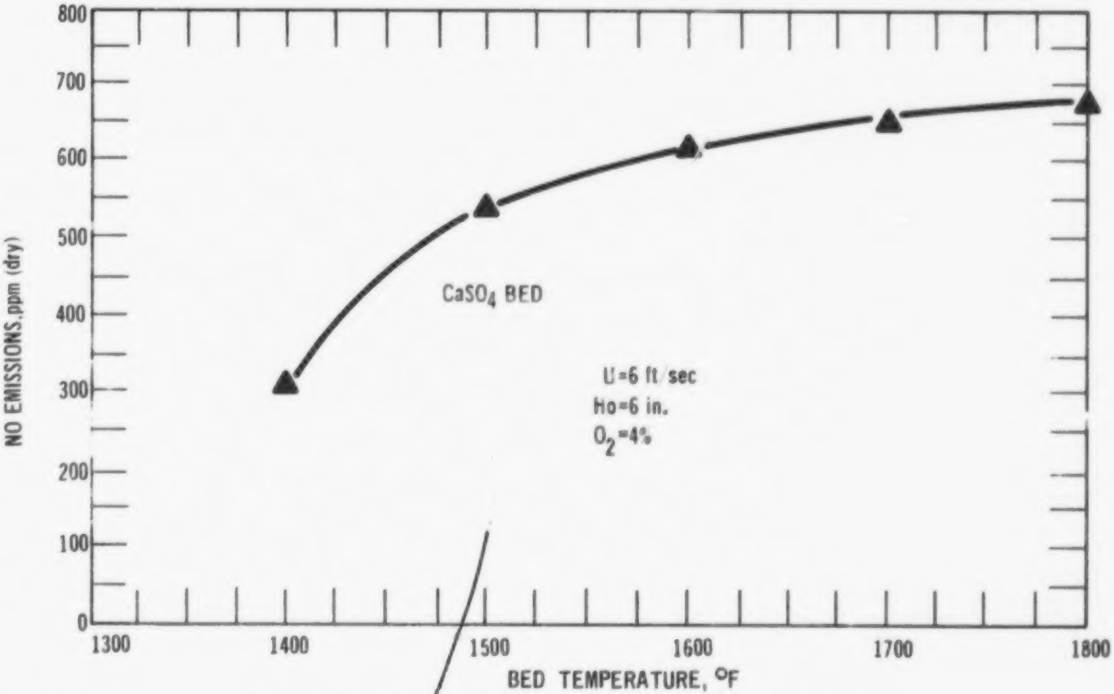


Figure 4. NO emissions as a function of bed temperature.

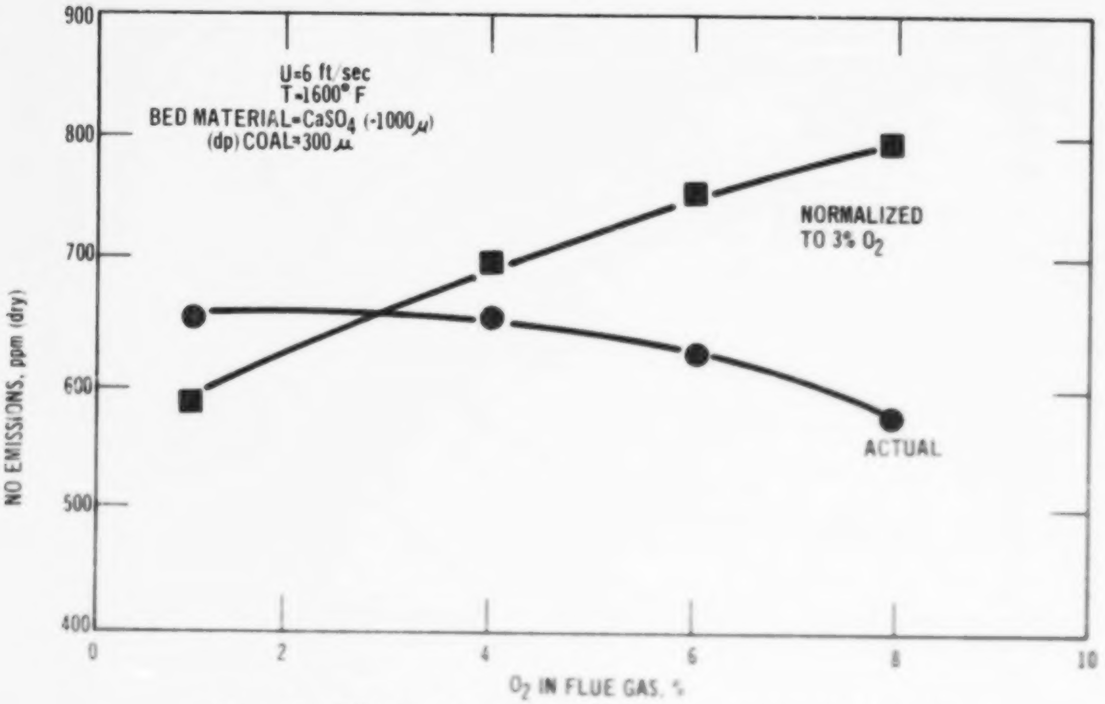


Figure 5. Effect of O_2 in flue gas on NO emissions ($CaSO_4$ bed).

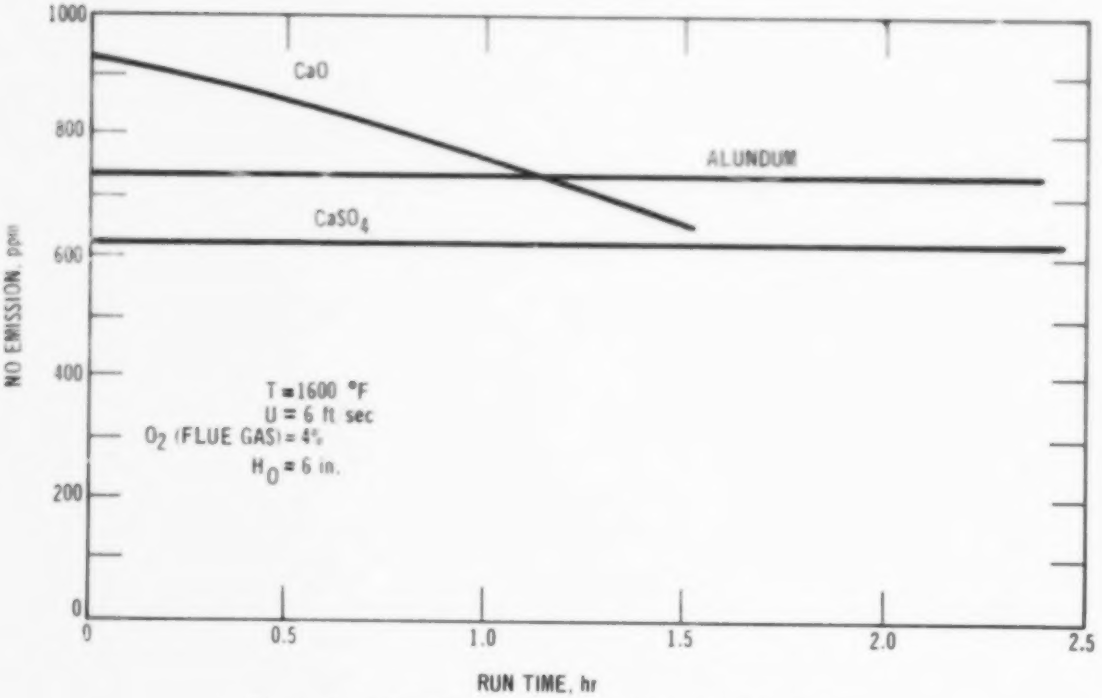


Figure 6. NO emissions using different bed materials.

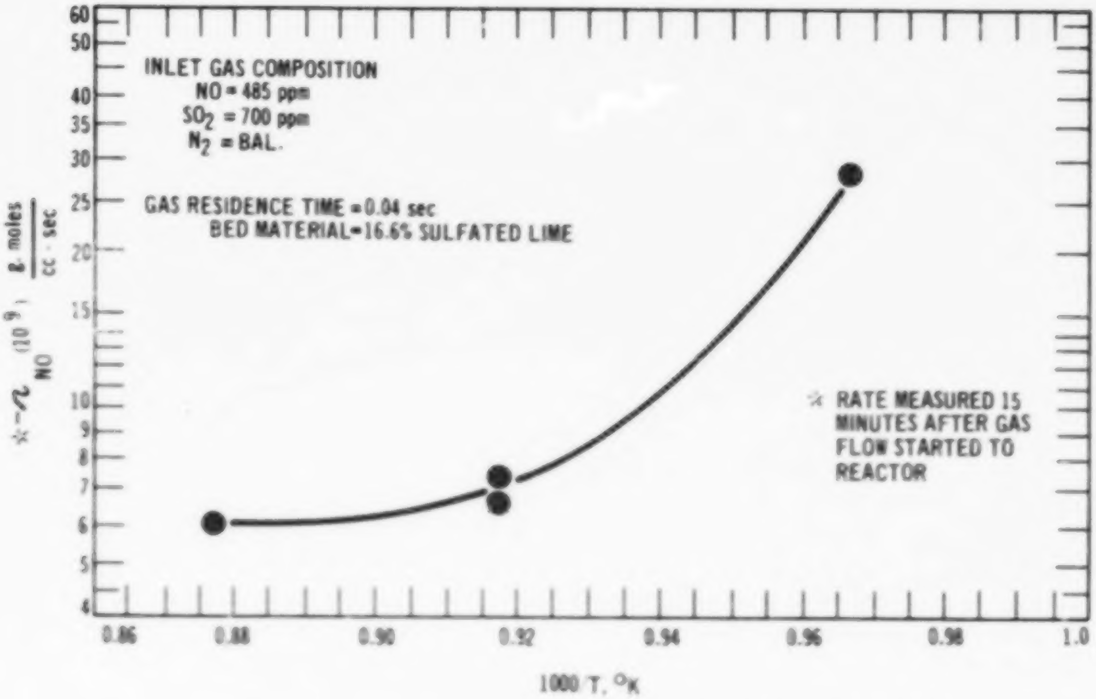


Figure 7. Temperature dependence of NO-SO_2-CaO reaction system.

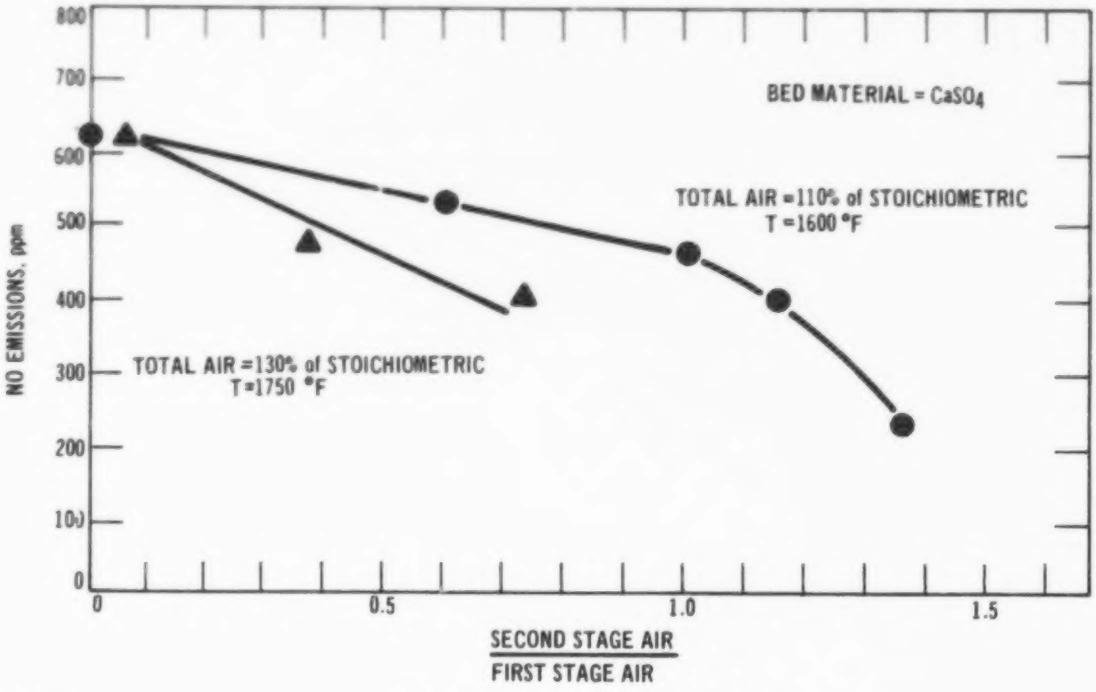


Figure 8. Staged FBC results.

3. COMBUSTION OF COALS IN FLUIDIZED BEDS OF LIMESTONE

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INTRODUCTION

The experimental work reported here was performed by the Bureau of Mines, Morgantown Energy Research Center, under contract to the Control Systems Division, Office of Research and Monitoring, Environmental Protection Agency. The phase of work assigned to the Bureau was to test various coals as fuel in fluidized beds of limestone, to compare sulfur retention, and to measure heat transfer with tubes immersed in the bed. The program involved testing five types of bituminous coal from high volatile A to low volatile, which varied in ash content from 8 to 24 percent and in sulfur content from 2 to 4 percent.

EQUIPMENT AND PROCEDURE

Figure 1 shows the 8-ft high combustor used in the tests. The bed was supported by a cone-shaped plate, perforated by 1/8-in. holes fitted with welded stainless steel 90° elbows to inject the fluidizing air axially and parallel to the cone surface. Water passing through a heat exchanger made of 3/4-in. pipe extracted heat from the combustion bed. Tests were performed at superficial fluidizing velocities of 3 ft/sec and 6 ft/sec. In the 6-ft/sec tests, an additional heat exchanger was installed to control the temperature of the gases leaving the combustor.

Figure 2 is a flow diagram of the system. Coal was metered by a screw conveyor, and then fed pneumatically near the base of the bed. Limestone was fed through the side of the combustor just above the bed by a screw conveyor. The bed level was maintained by periodically removing material from the bottom with a 3-in. screw conveyor. Combustion products were passed through two centrifugal separators for removal of most of the entrained solids, and then to a bag filter for final cleaning. Solids from the first cyclone could be reinjected into the combustion zone. Combustion gases were monitored continuously for O_2, CO_2, CO, and SO_2 by infrared analyzers except for O_2 which utilized a paramagnetic system. After each test the residue was removed from the bottom of the combustor by the screw conveyor.

Startup was accomplished in about two hours by burning natural gas in the combustor and then injecting coal mixed with limestone into the combustion chamber. A 2-ft bed was established at about 1200°F, after which the gas was shut off, and coal and limestone were fed at a rate that is compatible with the superficial air velocity and the designated run conditions.

Combustion Tests

Five types of coal were burned in beds of limestone to determine its effectiveness for retaining sulfur. The limestone was the type designated BCR 1359 (97 percent $CaCO_3$, Northern Virginia); various sizes of limestone were used in tests at 3 ft/sec but in tests at 6 ft/sec the limestone was sized to 1/4- by 3/16-in. The coals were crushed by a hammer mill to the range of sizes shown in Table 1. Typical analyses of the various types of coal are given in Table 2. These analyses varied somewhat throughout the test series because batches of the same coals were purchased at different times.

Table 1. TYPICAL SIZE RANGE OF COALS BURNED IN FLUID-BED COMBUSTION TESTS

Screen size, mesh (USS)	Weight percent
-1/4-inch + 20	41-53
-20 + 40	16-24
-40 + 100	15-20
-100 + 150	2-6
-150 + 200	2-5
-200	4-10

Combustion tests, generally of over 70-hr duration, were made at fluidizing velocities of 3 and 6 ft/sec. At 3 ft/sec, several tests were made with each coal; at 6 ft/sec, only one test was made with each coal. Results of the tests are given in Table 3. Figure 3 shows the effect of Ca/S mole ratio on sulfur retention in the bed. It should be noted (from Table 3) that for most of the tests at 3 ft/sec, material from the primary cyclone was recycled to the bed. At 6 ft/sec, recycle was possible only with one coal, hvbb, due to cooling of the bed by reinjection of the large volume of solids.

The data of Figure 3 generally show that for the coals burned at 3 ft/sec there appears to be a trend in which S removal increases rapidly to 90 percent as Ca/S is increased to approximately 2. Two of the tests,however, appear to deviate from this general pattern: hvab (A-5-L), Ca/S = 1.8, S removal = 73 percent; lvb (G-5-L), Ca/S = 2.0, S removal = 68 percent. The test with hvab (A-5-L) was one of the first tests made and was more cyclic in

Table 2. TYPICAL ANALYSES OF COALS BURNED IN FLUID-BED COMBUSTION TESTS

	hvcb (Ill. #6)	hvbb (Ind. #5)	hvab (Ohio)	hvab (W. Va.)	mvb	lvb
Proximate analysis, wt %						
Moisture	2.4	6.0	2.3	1.1	1.1	1.1
Volatile matter	33.8	35.5	39.8	33.1	19.8	17.3
Fixed carbon	55.6	50.6	49.7	58.0	62.4	69.9
Ash	8.2	7.9	8.2	7.8	16.7	11.7
Ultimate analysis, wt %						
Carbon	72.4	69.6	72.3	76.5	71.2	76.0
Hydrogen	5.1	5.2	5.5	4.8	4.4	4.0
Nitrogen	1.2	1.2	1.5	1.4	1.0	1.2
Sulfur	2.6	2.9	3.9	3.0	3.3	2.3
Oxygen	8.1	7.2	6.3	5.4	2.3	3.7
Heating value, Btu/lb	13,045	12,530	12,930	13,820	13,050	13,120

nature than the later tests. In the test with lvb (G-5-L), the SO_2 meter functioned only a part of the time, so the average SO_2 concentration is suspect. Thus, there is reason to believe that the average results reported for tests A-5-L and G-5-L are not representative.

The results from tests at 6 ft/sec show a slightly different pattern, but duplicate tests would have to be made to confirm this. In only one of these six tests, that with hvbb, material from the primary cyclone was recycled to the bed. Four of the remaining five tests without recycle indicated the S retention increases as Ca/S is increased, but not as rapidly as in the 3 ft/sec tests, and that Ca/S of 3 or more is required to retain approximately 90 percent of the sulfur in the bed. The test at 6 ft/sec with lvb does not fit the pattern, but no explanation can be offered. In the one test at 6 ft/sec when recycle was used, S removal was 89 percent at Ca/S = 2.1, closely approximating results from tests at 3 ft/sec when recycle was employed. Therefore, based on the few tests at 6 ft/sec, it is difficult to determine whether the reduced S removal was caused by the increase in fluidizing velocity or the absence of recycle.

Increasing the fluidizing velocity has the following effects:

1. Gas residence time in the bed is reduced.
2. Bed density is decreased which lessens gas-solids contact.
3. Higher rates of limestone are required resulting in a reduction of solids residence time.
4. Larger gas bubbles are formed which permit more bypassing of the SO_2.

All of the above effects of increasing the fluidizing velocity would tend to decrease S retention. In addition, since the entrained solids leaving the combustor likely contain some unreacted limestone, recycling of this material would be expected to improve S retention. Thus, the lower S retention at 6 ft/sec is probably caused by both the higher velocity and the absence of recycle.

Table 3. RESULTS OF FLUID-BED COMBUSTION OF COALS IN BEDS OF LIMESTONE, BCR-1359

	Type of coal/run number											
	hvcb				hvbb					lvb		
					Enos Mine		Blackfoot Mine					
	E-4	E-5	E-6	E-7	C-4	C-5	C-6	C-7	C-8	G-5	G-6	G-8
Duration, hr	84	79	72	50	87	84	73	70	84	84.5	60	46
Bed temperature, °F	1500	1510	1505	1520	1560	1510	1510	1520	1545	1535	1470	1530
Superficial velocity, ft/sec	3.0	3.0	3.0	5.9	3.1	3.0	3.0	3.0	6.2	2.7	3.0	6.0
Coal rate, lb/hr	52.9	51.0	49.5	72.7	36.9	33.8	34.0	31.7	67.4	31.1	36.3	87.6
Air/coal, scf/lb	95.5	98.8	101.5	140.2	137.1	148.0	148.2	161.6	154.4	143.5	138.8	112.4
Limestone rate, lb/hr	15.0	12.7	13.3	17.4	4.0	5.0	12.7	13.4	13.2	5.6	5.7	11.1
Sulfur in coal, wt %	3.7	3.7	3.7	4.1	4.0	4.0	3.1	2.9	2.9	2.8	2.3	2.6
Ca/S mole ratio	2.5	2.2	2.4	1.8	0.85	1.2	3.8	4.4	2.1	2.0	1.8	1.5
SO_2 in POC, ppm	163	416	461	1094	993	520	305	189	273	800	192	121
Sulfur removal, wt %	95.8	90.8	89.5	69.8	71.4	83.8	87.8	91.7	88.7	67.7	92.5	96.1
Recycle in use	Yes	Yes	Yes	No	Yes	Yes	Yes	Yes	Yes	Yes	Partial	No
Carbon utilization, %	-	89.8	92.7	75.6	98.8	97.1	97.3	92.5	96.3	85.3	88.6	65.0

Table 3 (continued). RESULTS OF FLUID-BED COMBUSTION OF COALS IN BEDS OF LIMESTONE, BCR-1359

	Type of coal/run number											
	hvab (W. Va.)				hvab (Ohio)			mvb				
	Humphrey Mine		Love-ridge Mine	Ire-land Mine								
	A-5	A-6	A-7	I-1	B-4	B-5	B-6	F-4	F-5[a]	F-6	F-7[a]	F-8[a]
Duration, hr	84	71	84	84	75	84	75	60.5	84	36	84	80
Bed temperature, °F	1525	1525	1525	1535	1580	1520	1560	1505	1490	1435	1495	1575
Superficial velocity, ft/sec	3.1	3.0	3.0	6.3	3.0	3.0	6.0	2.8	3.0	2.6	3.0	6.0
Coal rate, lb/hr	34.4	31.8	31.4	66.6	34.3	34.8	58.9	30.1	41.8	52.2	36.1	65.3
Air/coal, scf/lb	144.8	157.4	160.3	157.5	144.1	149.0	170.7	155.2	118.0	86.3	140.0	154.7
Limestone rate, lb/hr	4.4	3.9	9.7	13.8	4.3	5.9	15.2	4.6	8.0	10.0	9.3	15.2
Sulfur in coal, wt %	2.2	2.2	3.0	4.2	3.9	3.9	4.1	5.2	3.3	5.2	3.3	2.5
Ca/S mole ratio	1.8	1.7	3.4	1.5	1.0	1.4	2.0	0.9	1.6	1.2	2.2	2.9
SO_2 in POC, ppm	496	63	134	848	1003	194	594	2310	445	2036	196	293
Sulfur removal, wt %	73.1	96.3	93.6	73.9	69.4	94.1	80.0	42.7	88.5	72.4	93.9	86.3
Recycle in use	Yes	Yes	Yes	No	Yes	Yes	No	No	No	No	Partial	No
Carbon utilization, %	98.0	95.1	96.6	91.1	-	93.2	85.3	-	82.1	81.4	89.9	74.1

[a] Coal was air-table cleaned.

Results in Table 3 also show that when material from the primary cyclone is not recycled to the bed, carbon burnup decreases. This is even true at the lower velocity of 3 ft/sec. In a commercial boiler, if recycle was not used, the boiler would have to incorporate a method for increased carbon utilization such as the "carbon burnup cell" proposed by Pope, Evans and Robbins.[1] Results from tests at 3 ft/sec show carbon utilizations with recycle to range from about 90 to 99 percent. To consistently achieve acceptable burnup, i.e., more than 99 percent, recycle might not obviate the need for a burnup cell.

Heat Transfer

Heat transfer in a fluid-bed boiler is important in establishing the commercial potential of this combustion technique and would also be important in the design of fluid-bed boilers. In the combustor previously described, which contained a series of water-cooled U-tubes immersed in the bed, data were taken on one U-tube during the combustion tests. Values of U_i were calculated from the data and values for the water coefficient (h_i) were calculated using the Dittus-Boelter relationship. Values for the bed-to-tube coefficient (h_B) were then calculated by the relationship

$$U_i = \frac{1}{\frac{1}{h_i} + \frac{D_i x}{D_{av} k} + \frac{D_i}{D_o h_B}} \quad (1)$$

where:

U_i = overall heat transfer coefficient based on inside area of pipe

h_i = inside film coefficient, steam

h_B = outside film coefficient, fluid bed

D_i, D_{av}, D_o = inside, average, outside pipe diameters

x = pipe wall thickness

k = thermal conductivity of pipe.

Results are given in Table 4 for 18 tests at 3 ft/sec and six tests at 6 ft/sec. The results from tests at 3 ft/sec are generally consistent, except for two tests (B-5-L, C-7-L). Neglecting those two tests, the average bed-to-tube film coefficient is 67.5 Btu/hr-ft^2-°F. At 6 ft/sec, the average bed-to-tube coefficient was 32.3 Btu/hr-ft^2-°F.

Heat transfer also was investigated in another 18-in. diameter combustor that was operated to evaluate the performance of various alloy tubes. This combustor contained a steam-cooled tube bundle which passed horizontally through the fluid bed. Figure 4 shows the layout of the tube bundle. During three tests of approximately 500 hours each in duration, one at 3 ft/sec and two at 6 ft/sec, heat transfer to the various tubes was measured.

Heat transfer data from this steam-cooled tube bundle were examined via graphical interpretation of overall heat transfer coefficients. The overall heat transfer coefficient based on the inside area of the pipe is given by preceding equation. Since conditions in the fluid bed were essentially constant in each long-duration test, h_B should be essentially constant. Neglecting thermal expansion of the tubes, D_i, D_o, D_{av}, and x are constants for the tubes; k is constant for the various alloys over the temperature range of the alloys (k is somewhat higher for carbon steel, but the resistance term for the metal wall is so small that it is insignificant). Hence, the resistance terms for the metal wall and the outside film can be combined into one constant R_1

$$R_1 = \frac{D_i x}{D_{av} k} + \frac{D_i}{D_o h_B} \quad (2)$$

Thus, $$U_i = \frac{1}{\frac{1}{h_i} + R_1} \quad (3)$$

Table 4. HEAT TRANSFER RESULTS FROM WATER-COOLED U-TUBE

Test	Limestone size	U_i Btu/hr-ft^2-°F	h_{Bed} Btu/hr-ft^2-°F
	Fluidizing Velocity = 3 ft/sec		
A5L	1/4 in. x 10 mesh	71.2	75.5
A6L	3/8 in. x 3/16 in.	68.9	70.1
A7L	3/16 in. x 30 mesh	55.4	52.0
B4L	3/8 in. x 3/16 in.	64.0	60.9
B5L	8 x 28 mesh	36.1	31.9
C4L	1/4 in. x 10 mesh	61.3	61.8
C5L	3/8 in. x 3/16 in.	60.8	60.9
C6L	8 x 28 mesh	62.4	61.3
C7L	1/4 in. x 3/16 in.	31.2	26.7
E4L	8 x 28 mesh	69.6	67.7
E5L	8 x 28 mesh	69.1	68.3
E6L	3/16 in. x 30 mesh	66.0	64.3
F4L	3/8 in. x 3/16 in.	68.5	70.1
F5L	3/8 in x 3/16 in.	79.1	83.5
F6L	3/8 in. x 3/16 in.	67.7	70.7
F7L	8 x 28 mesh	65.1	66.0
G5L	3/8 in. x 3/16 in.	70.8	73.4
G6L	8 x 28 mesh	68.8	72.7
			Avg. 67.5[a]
	Fluidizing Velocity = 6 ft/sec		
B6L	1/4 in. x 3/16 in.	36.9	32.0
C8L	1/4 in. x 3/16 in.	39.9	34.9
E7L	1/4 in. x 3/16 in.	37.6	32.3
F8L	1/4 in. x 3/16 in.	39.9	34.7
G8L	1/4 in. x 3/16 in.	34.4	29.5
I1L	1/4 in. x 3/16 in.	34.9	30.1
			Avg. 32.3

[a] Neglecting two abnormally low values.

or

$$\frac{1}{U_i} = \frac{1}{h_i} + R_i \quad . \qquad (4)$$

For a gas or vapor flowing inside smooth, circular pipe in turbulent flow, the film coefficient h_i is given by a number of relationships of the type:

$$h_i = \frac{nG^{0.8}}{D_i^{0.2}} \qquad (5)$$

where:

n = a constant that depends on the physical properties of the fluid

G = mass velocity of the fluid

D_i = inside diameter of the pipe.

For steam over the temperatures in these tests, n can be considered constant, and D_i is constant for all the tubes. Therefore,

$$h_i = CG^{0.8}$$

and $$\frac{1}{U_i} = \frac{1}{CG^{0.8}} + R_1$$

where: C is a constant.

Data from each tube were used to calculate Q, the rate of heat transfer. Values of Q were then substituted into the formula $Q = U_i A \Delta t$ to obtain values for U_i. A plot of $1/U_i$ (as ordinate) versus $1/G^{0.8}$ gives a straight line with a slope of 1/C. The vertical intercept of this line, b, represents R_1, the sum of the resistance of the metal wall and the outside (fluid bed) film.

Figures 5 and 6 show the graphical interpretations of the overall coefficients of heat transfer. The linear correlation for the first test (Figure 5) has a vertical intercept of 0.0127, and a calculated fluid-bed film coefficient (h_B) of 63.4 Btu/hr-ft^2-°F. The correlations for second and third tests (Figure 6) were combined since both tests were made at 6 ft/sec and with -1/4 in. + 3/16 in. limestone. In this latter case, the vertical intercept is 0.0185, and a calculated fluid bed film coefficient (h_B) of 43.3 Btu/hr-ft^2-°F.

The graphical interpretations appear reasonable. Results from the test at 3 ft/sec with -8 + 30 mesh stone gave a bed coefficient of 63; results from the two tests at 6 ft/sec with beds of -1/4 in. + 3/16 in. stone gave a bed coefficient of 43. The differences between the two values were caused by differences in particle size and fluidizing velocity. The location of data points from the second test suggests there is a difference in heat transfer between the top and bottom rows; data from the first and third tests do not appear to support this.

At the same superficial fluidizing velocity, results from the steam-cooled and water-cooled exchangers were expected to be comparable. At 3 ft/sec, the agreement was quite good,with bed coefficients of 63.4 for the steam-cooled tubes and 67.5 for the water-cooled U-tube. At 6 ft/sec, the agreement was not as good: 43.3 for the steam-cooled tubes and 32.3 for the water-cooled U-tube.

CONCLUSIONS

At a fluidizing velocity of 3 ft/sec, and when fines from the primary cyclone are recycled to the bed, S retention by a limestone bed increases rapidly to 90 percent as Ca/S is increased to approximately 2. At 6 ft/sec and without recycle, it appears that Ca/S must be at least 3 to retain 90 percent of the S in the bed.

Carbon burnup was too low for commercial boiler operation when recycle was not employed, regardless of the fluidizing velocity. Even when recycle was used at the lower velocity of 3 ft/sec, carbon burnup might not be commercially acceptable so that a separate burnup cell would be required.

At 3 ft/sec fluidizing velocity, using beds ranging in size from 8 x 28 mesh to 3/8- x 3/16-in., heat transfer coefficient from the bed to a tube immersed in the bed was 60 to 70 Btu/hr-ft^2 -°F. At a velocity of 6 ft/sec and with beds of 1/4- x 3/16-in. particles, the coefficients to steam-cooled and water-cooled tubes were 43 and 32, respectively.

REFERENCE

1. Bishop, J.W., E.B. Robinson, E. Erlich, A.K. Jain, and P.M. Chen. Status of the Direct Contact Heat Transferring Fluidized Bed Boiler. (Presented at Winter Annual Meeting, ASME, New York. Paper Number 68-WA/FU-4. December 1-5, 1968.)

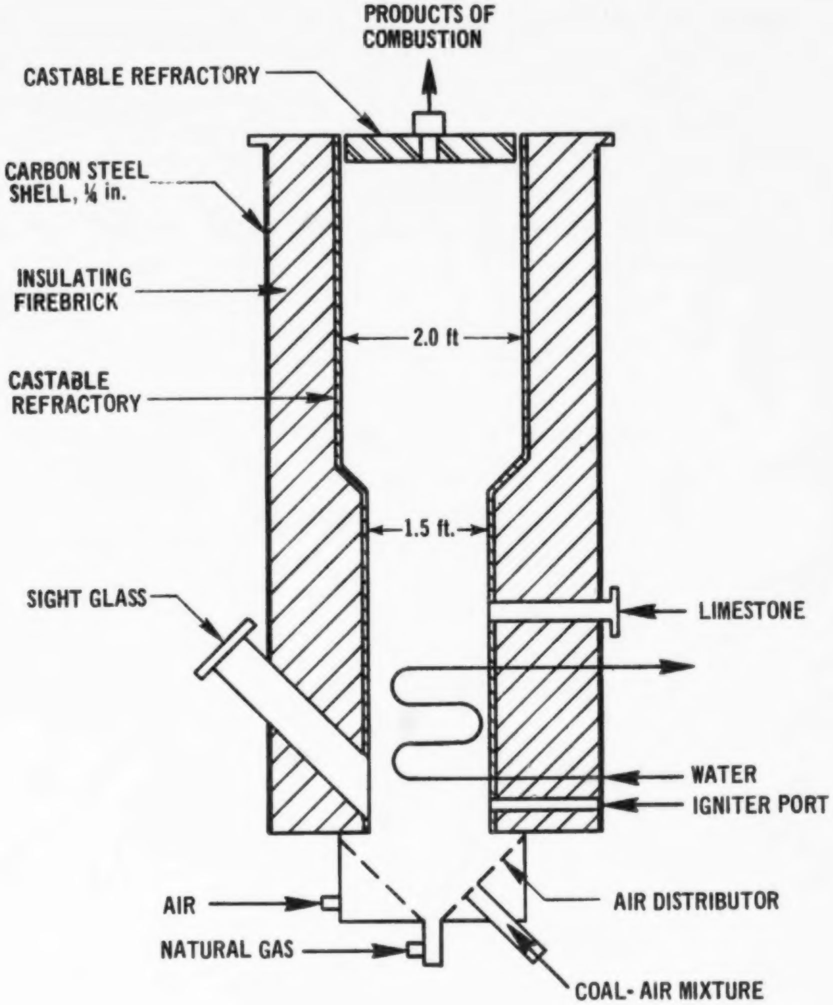


Figure 1. Fluid-bed combustor.

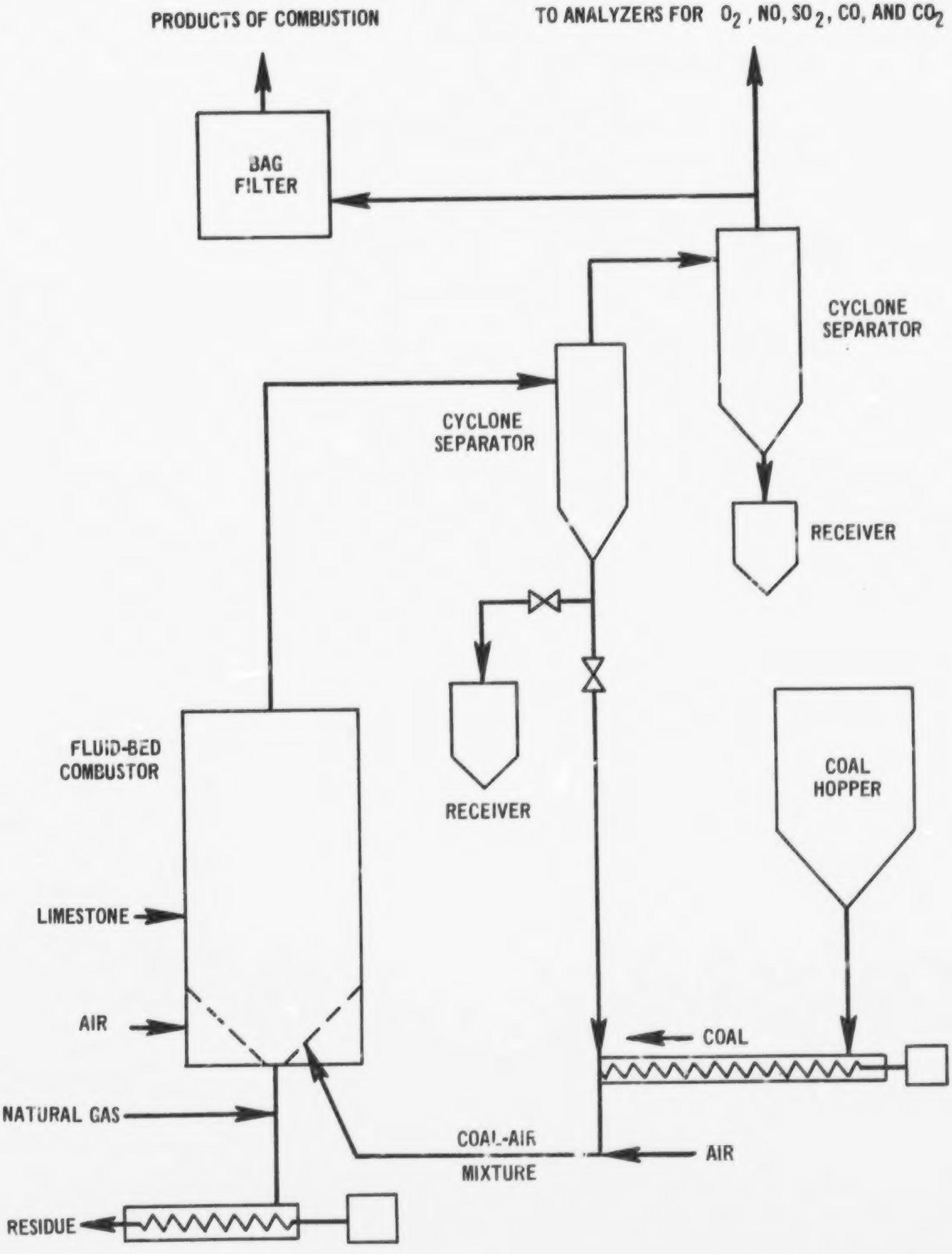


Figure 2. Flowsheet for fluid-bed combustion system.

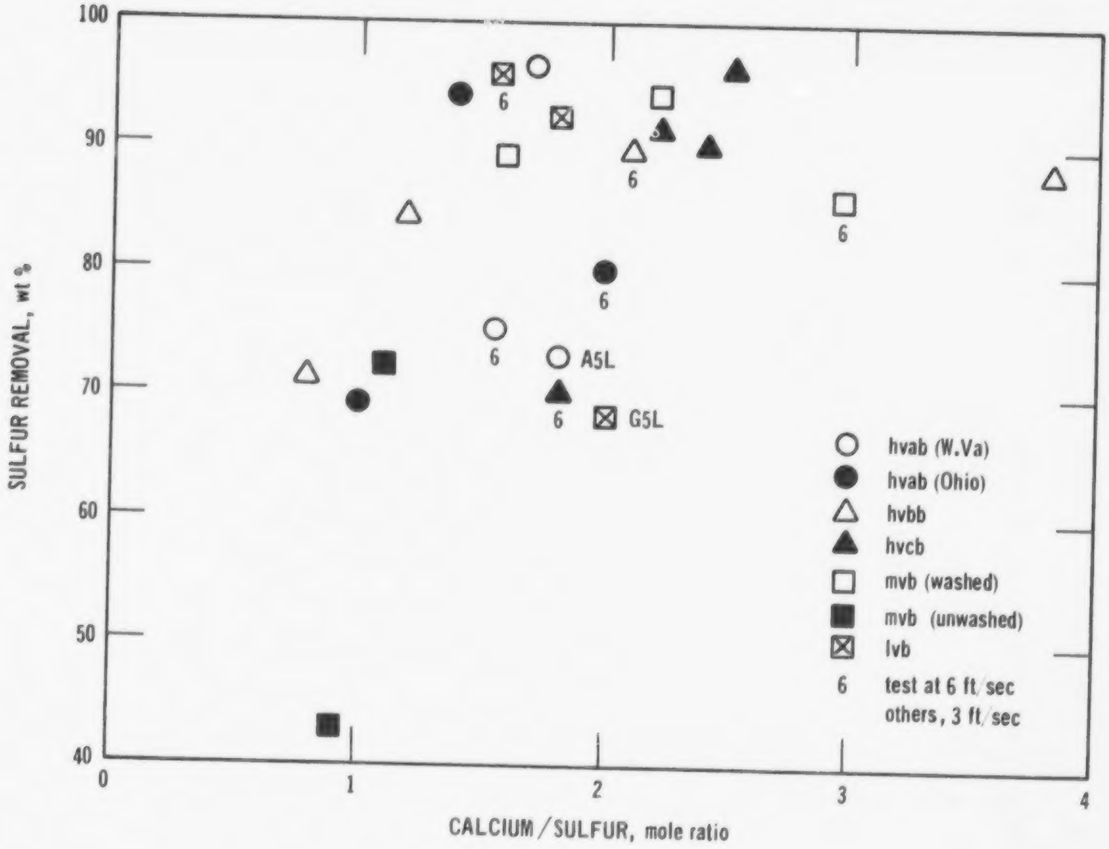
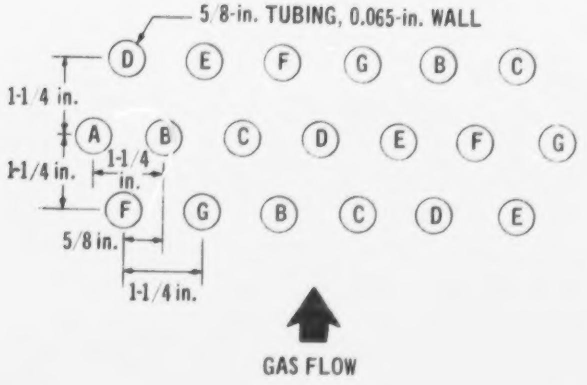


Figure 3. Effect of Ca/S mole ratio on sulfur removal.



TUBE	MATERIAL
A	SEAMLESS CARBON STEEL
B	WELDED 340 STAINLESS STEEL
C	SEAMLESS 316 STAINLESS STEEL
D	SEAMLESS 410 STAINLESS STEEL
E	SEAMLESS 446 STAINLESS STEEL
F	SEAMLESS 304 STAINLESS STEEL
G	WELDED 316 STAINLESS STEEL

Figure 4. Arrangement of steam-cooled tube bundle.

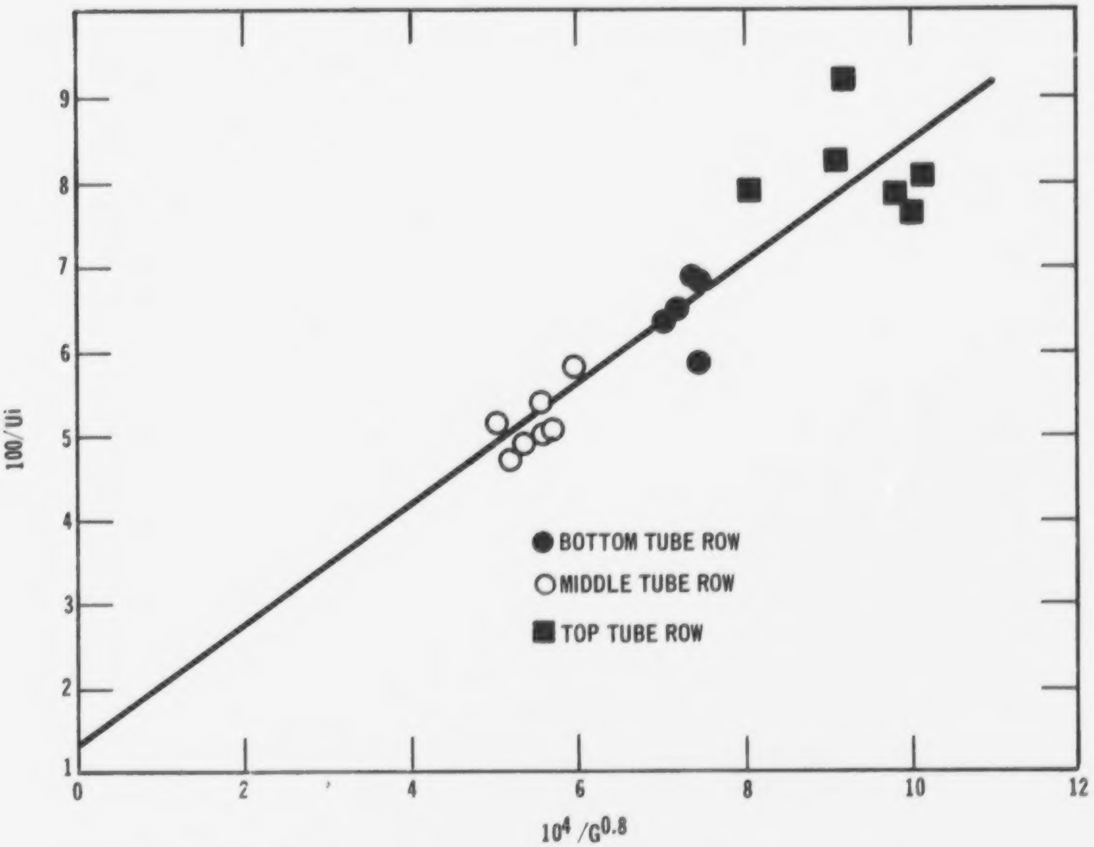


Figure 5. Graphical analysis of overall heat transfer coefficients: 3 ft/sec fluidizing velocity, -8+30 mesh limestone.

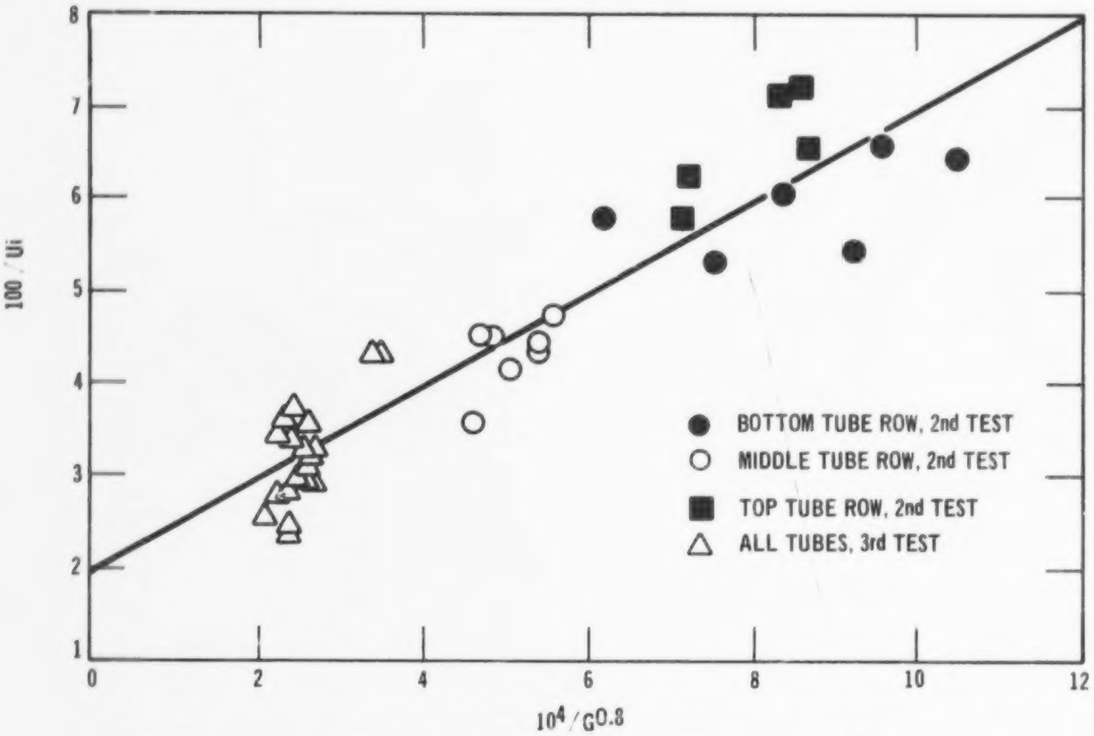


Figure 6. Graphical analysis of overall heat transfer coefficients: 6 ft/sec fluidizing velocity, - 1/4 in. +3/16 in. limestone.

4. THE REDUCTION OF EMISSIONS OF SULPHUR OXIDES AND NITROGEN OXIDES BY ADDITIONS OF LIMESTONE OR DOLOMITE DURING THE COMBUSTION OF COAL IN FLUIDISED BEDS

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INTRODUCTION

It has been known for a number of years, at least since the First International Conference on Fluidised-Bed Combustion held in the autumn of 1968, that additions of limestone or dolomite to fluidised-bed combustors could materially reduce the proportion of the sulphur which was emitted in the flue gases as sulphur dioxide.

As research work progressed, both in the U.S.A. and the U.K., it became apparent that there were significant and unexplained differences between results obtained at different establishments under apparently similar conditions. At that time the National Coal Board (NCB) had in operation the most comprehensive range of fluidised-bed combustors available. In May 1970, therefore, the National Coal Board and the Environmental Protection Agency agreed to jointly finance a considerable experimental programme designed (1) to establish the causes of some of the anomalies in the extant data, (2) to establish, within the range of the rigs available, the effects of scale, and (3) to systematically investigate some of the many variables effecting sulphur retention in fluidised combustion beds.

The programme was scheduled to cover a period of 12 months and involved the following work at the NCB's Leatherhead and Cheltenham laboratories.

1. Experiments on a number of pilot-scale combustors to measure the effect on emission of sulphur, nitrogen oxides, and particulates of a selected range of process conditions; e.g. coal type; the quantity, size and type of additive (limestone/dolomite) fed to the combustor to retain sulphur; combustion conditions as regards temperature, pressure, and fluidising velocity; plant scale; and design features such as bed depth and the recycling of incompletely reacted fuel and additive.
2. Experiments on selected pilot-scale combustors to assess the extent to which the addition of limestone or dolomite to coal in a fluidised bed in a large test rig influences the corrosion, erosion, and deposit formation on specimens representative of typical evaporator, superheater, and reheater tube metals.
3. Laboratory scale experiments to characterise the coals and additives used.
4. Development of a mathematical model to assist in correlating the factors which influence the pollution control characteristics of a fluidised combustion system.

THE SCOPE OF THE RESEARCH PROGRAMME

The main objectives of the research programme were:

1. To assess the effectiveness of the fluidised-bed combustion process, with and without the addition of limestone or dolomite, towards the reduction of SO_2 emission; to show those operating parameters that significantly affect the attainment of the immediate target emission (300 ppm v/v); and to indicate how these data may affect plant design.
2. To gather data, over the same range of operating conditions, on the levels of NO_x emission that occur during the combustion process when the SO_2 is partially absorbed by added limestone or dolomite.
3. To measure the particulates elutriated from the fluidised-bed combustor in order to provide data for the design of a particulates removal system which will reduce atmospheric emissions to an acceptable level.
4. To contribute towards an understanding of the way in which the porous properties of limestone or dolomite affect SO_2 retention under the conditions prevailing in a fluid-bed combustor, and to develop a simple method of classifying limestones and dolomites according to their utility for SO_2 retention in the fluidised-bed combustion process.
5. To develop a mathematical model of the retention of SO_2 in a fluid-bed combustion system to allow the performance of new plant, with respect to SO_2 emission, to be predicted at the design stage from the design and other basic data.
6. To study corrosion of typical steels used in boiler construction when immersed in a fluid-bed burning coal, both with and without the addition of limestone/dolomite.

The research programme to meet these objectives was organised into eight tasks, at various plants or locations as follows:

1. To compare the performance of the 36-in. rig with that of the 6-in. rigs at C.R.E. and Argonne, and to extend the range of operating conditions for which experimental data are available (36-in. combustor, CRE).
2. To obtain, for operation under pressure, data on the emission of sulphur and nitrogen oxides and on corrosion/deposition of boiler metal and turbine blade specimens (48- x 24-in. pressurised combustor, BCURA).
3. To carry out long-term tests to assess the effect of limestone addition on corrosion of evaporator, superheater, and reheater metals immersed in the fluid bed (27-in. combustor, BCURA).
4. To obtain data on corrosion of evaporator, superheater, and reheater materials for lower fluidising velocities (12-in. combustor, CRE).
5. To obtain data on sulphur retention for a range of coals and limestones, in particular to allow comparison to be made with the 6-in. rig at Argonne (6-in. combustor, CRE).
6. To complete the development of a mathematical model of sulphur retention, to compare its predictions with the results of laboratory and rig experiments, and to up-date it as appropriate (mathematical work, BCURA).
7. To investigate the distribution of sulphur in a range of coals and in the residue from the rigs (laboratory work, CRE/BCURA).
8. To investigate the pore structure and related factors that affect sulphur retention by lime (laboratory work, BCURA).

The features of the combustors used, the range of operating conditions explored, and the aggregate number of test hours accomplished are shown in Tables 1 and 2. It will be noted that experiments were carried out at combustion pressures up to 5 atmospheres absolute, fluidising velocities up to 11 ft/sec, bed temperatures up to 1680°F, using four different coals, three limestones, and two dolomites; the test running time totalled 5300 hours.

MATERIALS USED

The experimental work in the programme was carried out using four coals, three limestones, and two dolomites. Typical analyses of the materials are given in Tables 3 and 4.

DISCUSSION OF THE RESULTS

There were considerable differences, both in size and geometry, between the various rigs used in the programme. For instance:

1. Rigs were either circular or rectangular in cross section and ranged in area from 0.2 to 8.0 ft^2.
2. The geometries of cooling surfaces within the bed ranged from deep banks of closely spaced 1-in. diameter tubes to relatively shallow banks of widely spaced 2.4-in. diameter tubes.
3. The area of the cross section served by a single coal feed point varied from 0.2 to 4.5 ft^2.
4. Some combustors had only internal fines recycle systems, some had both internal and external recycle systems, and others had only external recycle systems. Results referred to as being without recycle are from rigs with only external, and hence controllable, recycle systems.
5. In some combustors the walls of the bed and freeboard were uncooled; in others they were cooled throughout.

Despite those differences it was found that geometry as such was not a variable; the whole body of the results could be discussed in terms of the process variables.

Table 1. MAIN FEATURES OF THE PILOT-SCALE COMBUSTORS

Feature	Location				
	BCURA, Leatherhead		CRE, Cheltenham		
Designation	27 in.	48 in. x 24 in. (pressurised)	36 in.	12 in. (corrosion)	6 in.
Bed cross section	27 in. dia	48 in. x 24 in.	36 in. x 18 in.	12 in. x 12 in.	6 in. dia
Bed depth, ft	1.5 - 2.0	3.5 - 4.0	2 - 7	2	2 - 3
Operating pressure, atm abs.	1	up to 5	1	1	1
Fluidising velocity, ft/sec	6 - 11	2	2 - 8	3	2 - 3
Coal rate, lb/hr	200 - 300	300 - 500	75 - 300	20 - 25	4 - 6
Total running hours	2150	430	1000	1100	600

Table 2. THE VARIABLES EXPLORED

Operating variables	Pilot-scale combustor(s)	
	Non-pressurised	Pressurised
Coal	U.S. Pittsburgh U.S. Illinois U.K. Welbeck U.K. Park Hill	U.S. Pittsburgh U.K. Welbeck
Ash content, %	12 - 18	13 - 18
Volatile matter, %	37 - 47	30 - 41
Sulphur, %	1.3 - 4.4	1.3 - 3.1
Chlorine, %	0.1 - 0.6	0.1 - 0.6
H_2O as fed, %	1 - 10	1 - 6
Ash fusion, °F	1800 - 2600	2100 - 2600
Size	-1/8 in. and -1/16 in.	-1/16 in.
Bed depth, ft	1.5 - 7	3.5 - 4
Temperature, °F	1420 - 1680	1470
Fluidising velocity, ft/sec	2 - 11	2
Excess air, %	-12 to +29	11 to 33
Recycle of cylone fines	Zero, partial, full	Partial
Additive	U.S. Limestone 18 U.S. Limestone 1359 U.K. Limestone U.S. Dolomite 1337	U.S. Limestone 18 U.S. Dolomite 1337 U.K. Dolomite
Ca/S mole ratio	0 - 6	0 - 3

The order in which the operating variables are commented upon takes into account both their relative importance and some of the interactions; e.g., through their effect on gas and solids residence times.

Pittsburgh coal (3 percent sulphur) was used with either Limestone 18 or Dolomite 1337 in the majority of experiments. While it is believed that most of the comments in the following statement of the main findings apply to other coals and limestones or dolomite, they refer primarily to these materials unless otherwise stated.

Ca/S Mole Ratio: The SO_2 is reduced asymptotically to zero as the feed rate of additive to the fluidised bed is increased. The percentage SO_2 reduction obtained at a given operating condition is a function of the mole ratio of added calcium to sulphur in coal; it is almost independent of the sulphur content of the coal, since the reaction is approximately first order with respect to SO_2 concentration. Clearly, in order to obtain a specified concentration of SO_2 in the off-gas when burning a coal of high sulphur content, it is necessary to achieve a higher percentage SO_2 reduction by using a higher Ca/S mole ratio.

For a given coal the lowest values of the Ca/S mole ratio were required at (1) low fluidising velocities (i.e. 2 to 3 ft/sec), (2) a bed

Table 3. TYPICAL ANALYSES OF COALS USED

Analysis	Coal			
	Illinois	Pittsburgh	Park Hill	Welbeck
Proximate analysis				
Total moisture, % a.r.	9.8	1.6	2.1	4.2
Ash, % a.r.	11.8	13.5	16.5	18.2
Volatile matter, % a.r.	46.6	41.1	39.2	38.3
Ultimate analysis				
Carbon, % d.b.	67.8	71.7	68.2	67.5
Hydrogen, % d.b.	4.5	4.5	4.4	4.3
Nitrogen, % d.b.	1.3	1.4	1.3	1.5
Sulphur, % d.b.	4.4	2.8	2.5	1.3
Oxygen + errors, % d.b.	8.5	4.4	5.3	5.1
Chlorine, % d.b.	0.2	0.1	0.1	0.6
Calorific value (d.a.f.), Btu/lb	14,300	15,100	14,750	14,400
Swelling number	4-1/2	8	1	1
Gray King coke type	D	G9	D	C
Ash analysis				
CaO, %	10.1	8.0	2.2	1.8
MgO, %	1.0	1.3	1.7	1.4
Na_2O, %	1.7	0.7	0.8	1.8
K_2O, %	1.8	1.6	3.6	3.2
SiO_2,%	40.8	45.8	46.0	57.5
Size (as received)	-1/4 in.	- 1/4 in.	-1-1/2 in.	-1-1/2 in.

Table 4. TYPICAL ANALYSES OF LIMESTONES AND DOLOMITES

Component	Dolomite 1337	U.K. Dolomite	Composition, % Limestone 18	Limestone 1359	U.K. Limestone
CaO	28.9	29.3	45.7	55.7	55.4
MgO	22.9	21.5	1.4	0.3	0.3
$H_2O + CO_2$	47.4	46.3	36.6	43.6	43.5
SiO_2	0.5	-	13.6	0.5	0.7
Fe_2O_3	0.2	-	0.3	0.1	0.1
SO_3	-	0.1	-	-	-
Total	99.9	97.2	97.6	100.2	99.8

temperature of around 1500°F, and (3) when most of the fines larger than about 10 μm were recycled. See Figure 1.

Bed Temperature: The optimum bed temperature was between 1400°F and 1600°F. The level of SO_2 emission and the change in emission with change of temperature on each side of the optimum appeared to depend on the type of additive used and to some extent on the Ca/S ratio employed. The increase in emission on either side of the minimum tended to be greater at low than at high Ca/S ratios; i.e. under conditions where the fraction of calcium sulphated was higher. The optimum temperature was found to be 1500-1550°F for limestone additive and 1400°F-1500°F for the dolomite. The data suggest that maintaining the same level of sulphur emission (e.g., 85 percent sulphur retention) at, for example, 100°F above the optimum, would involve increasing the Ca/S ratio by a factor of about two. The effect of changing bed temperature was not investigated on the pressure combustor.

The rapid increase in sulphur emission at bed temperatures above about 1550°F is unexpected from laboratory measurements of the reaction rate between CaO and SO_2. Since the effect of temperature appears to be reversible (i.e., the SO_2 reduction reverts to a high value as soon as the bed temperature is reduced) it cannot be accounted for by irreversible factors such as sintering or slag formation at the particle surface. One tentative explanation postulates that an oxygen-containing species (e.g., hydroxide ions derived from traces of water, which are known to be difficult to remove) is involved in the conversion of $CaSO_3$ to $CaSO_4$. It is possible that above the optimum temperature the hydroxide ions become more mobile and hence less able to participate in the reaction.

At low temperatures (i.e., below 1350°F) sulphur retention with dolomite was higher than with limestone, because of the lower calcination temperature of the $MgCO_3$ in dolomite which leads to the development of pore structure below the temperature of calcination of the $CaCO_3$. See Figure 2.

Fluidising Velocity: Increase in fluidising velocity resulted in an increase in sulphur emission. An empirical correlation was derived and is reported later in the summary. Increase in velocity without any compensating action results in reduction in both gas and solids residence times. To maintain the same sulphur retention (e.g., 85 percent, at 8 ft/sec as at 2 ft/sec fluidising velocity) the Ca/S mole ratio (at a bed temperature of 1500°F and without recycle) would have to be increased from about 2 to about 4 (Figure 3).

Bed Height: Increase in bed height usually resulted in a reduction in SO_2 emission. An empirical correlation is reported later in the summary. In principle it should be possible to counteract the adverse effect of increasing velocity by a proportionate increase in bed height. At atmospheric pressure the attendant increase in pressure loss for other than a small increase in bed height could be prohibitive. In addition, because the tube bank required would occupy only a part of the bed height, the effectiveness of increasing bed height may be reduced by the formation of large gas bubbles. The effect of bed height in super-charged boilers is potentially of greater significance. Here the deep banks of close packed tubes may assist in breaking up large gas bubbles and hence may improve the contact between gas and solids. Further, the increase in pressure loss due to increasing bed height is less important under pressure. See Figure 4.

Fines Recycle: A high proportion of the additive is elutriated from the bed before being fully utilised. By efficient recycle of fines larger than 10 μm, to the bed, SO_2 reduction was increased significantly; e.g., from 73 to 99 percent at a fluidising velocity of 2 ft/sec and a Ca/S mole ratio of 1.6.

Operating Pressure: The effect of operating pressure on SO_2 reduction was negligible when dolomite was used as an additive. This is to be expected with a reaction which is first order with respect to the partial pressure of

SO_2. With limestone as an additive, the reduction obtained at 5 atm was appreciably lower than with dolomite or with limestone at atmospheric pressure. This was also to be expected, since at 1470°F, calcination of limestone to give a porous structure would not occur at operating pressures above 2 atm. Penetration of the particle by SO_2 would therefore be difficult and only a surface layer of sulphate would form. It was found, however, that the performance of the limestone was better than this reasoning would imply; it suggests that the exposure of fresh surface by attrition plays a significant role. Nevertheless, from the point of view of both the Ca/S mole ratio and the total quantity of additive required to attain a target level of sulphur retention, dolomite was superior to limestone. To retain 85 percent of the sulphur, for example, the estimated Ca/S mole ratios for dolomite and limestone were 1.1 and 3.25 respectively; the estimated quantities of additive were 7.6 lb and 12 lb per lb of sulphur removed, respectively.

Particle Size: For coarsely crushed limestone the percentage SO_2 reduction increased when the particle size of limestone was reduced; this effect was probably due to the consequent increase in available reaction surface. On the other hand, with dolomite there was no effect of particle size, suggesting that access to internal surface is not a limiting factor for dolomite.

With additive ground to -125 μm or -150 μm, it was found that the fluidising velocity had a profound effect on SO_2 reduction. Whereas at low velocity (3 ft/sec) the fine additive improved SO_2 reduction; the reverse was true at high velocity (8 ft/sec). The data of Pope, Evans and Robbins suggest that, with a Ca/S mole ratio of 2.6 in beds 10 in. deep fluidised at 12 ft/sec, limestone 1359 gave 80 percent SO_2 reduction when ground to 44 μm, and 60 percent reduction when ground to -74 μm. Evidently the SO_2 reduction is very sensitive to the size of finely ground particles, so the Pope, Evans and Robbins data are at least qualitatively consistent with those for -150 μm limestone from the present study. Limestone ground to -150 μm may have too short a residence time at high velocity to achieve a high degree of sulphation; superfine material will become highly sulphated, since its residence time will not be markedly less than that of -150 μm material. However, superfine material may cause serious gas-cleaning problems.

Type of Additive: The type and source of additive affects the reduction in SO_2 that can be achieved. At atmospheric pressure Limestone 18 was the most effective additive on both molar and weight bases; the least effective on a mole basis was Limestone 1359, and on a weight basis Dolomite 1337. To achieve the same level of retention with the poorer Limestone 1359 as with the Limestone 18 would require an increase of up to 100 percent in the Ca/S mole ratio. As mentioned previously, for operation under pressure both the dolomites were superior to Limestone 18 on weight and molar bases.

An important finding from the point of view of simplifying prediction of suitability of stones was that measurements made at room temperature and at combustor temperatures showed the same accessibility of the structure to gases of similar molecular size to SO_2. Temperature cycling (as may occur in some plant designs when elutriated particles are recirculated) does not affect the pore structure significantly from the point of view of SO_2 uptake. An empirical reactivity test was considered to be the most economic method for classifying stones. For limestones the results of laboratory experiments give pessimistic predictions of plant performance. These results are thought to be because the tests do not take into account the beneficial effect of attrition in the combustor which results in removal of the sulphated surface layer. The effect of attrition was particularly important for limestones in the pressurised combustor and for Limestone 1359 at atmospheric pressure. For dolomite, access to the internal surface of the particles does not appear to be a limiting factor.

Whereas thermal losses are incurred from the sensible heat requirement and the heat of calcination of an additive, the heat of sulphation represents a thermal gain. Up to a Ca/S mole ratio of about 2 using limestone (sufficient to retain 85 percent of the sulphur of a 3 percent sulphur coal under good operating conditions), it is estimated that the heat of sulphation will counterbalance the sensible heat requirements and heat of calcination. With dolomite, however, the net thermal loss would be about 1-1/2 percent of the coal heat input. Under pressure calcination of $CaCO_3$ is inhibited, and the thermal loss incurred by using dolomite would be negligible for Ca/S mole ratios up to 2.

Type of Coal: The most important coal property in this context is the sulphur content, which determines not only the quantity of additive required for a given Ca/S mole ratio, but also the percentage SO_2 reduction (and hence Ca/S mole ratio needed) to meet set limits of SO_2 emission. Since the SO_2 absorption reaction is first order with respect to SO_2 concentration, it could be expected that the same relationship between percentage reduction and Ca/S mole ratio would hold for all coals irrespective of the sulphur content. However, the experimental results showed that, for a given Ca/S mole ratio, similar SO_2 reductions were obtained for three of the coals, but the reductions were up to 15 percent higher with Welbeck coal. Differences in the rate of sulphur release have been found between coals and might partly account for differences in performance. A more likely explanation of the higher SO_2 reduction with Welbeck coal is its low sulphur content, which had the consequence that additive was fed at a lower rate and hence had a longer residence time. This could have resulted in the higher degree of sulphation, particularly if particle attrition was an important effect.

Plant Design: As mentioned earlier, it was concluded that despite the difference of scale and design over the range of combustors used there was no significant difference between the SO_2 reductions obtained in different combustors with the same operating conditions. Nevertheless, direct application of the present results to combustors of commercial size requires some caution. Some factors which might alter the SO_2 emission, such as the depth of the tube bank, have already been mentioned. Further, observation of a radial distribution of SO_2 concentration in the freeboard of one of the larger pilot plants suggests that coal feed spacing, if greater than that used in the pilot-plants, may assume significance in commercial boilers.

Mathematical Model and Correlation of Data: The mathematical model has been developed to give fairly satisfactory prediction of the consequence of changing some operating conditions. Additional development is needed: (1) to take further account of attrition of additive and (2) to extrapolate the results to combustors that differ significantly from the present pilot plants. The model in its present form has not been useful in correlating the experimental data. However a number of empirical correlations have been derived as follows.

There is an approximately exponential relationship between the SO_2 reduction and the Ca/S mole ratio of the form

$$R = 100\,[1 - \exp(-MC)] \qquad (1)$$

where: R = percentage SO_2 reduction
C = Ca/S mole ratio
M = empirical constant depending on the coal, limestone, and operating conditions.

The effect of fluidising velocity on SO_2 reduction may be approximately correlated by

$$A = X_1/V \qquad (2)$$

where: A = absorption ratio, defined as R/(100-R)
V = fluidising velocity
X_1 = empirical constant depending on the Ca/S mole ratio and other operating conditions.

The effect of bed height on SO_2 reduction may be approximately correlated by

$$A = X_2 H \qquad (3)$$

where: H = bed height
X_2 = empirical constant depending on the Ca/S mole ratio and other operating conditions.

Emission of Nitrogen Oxides

Emission of NO_x from the pressure combustor (50-200 ppm) was significantly lower than from the non-pressurised combustors (300-600 ppm). All the combustors produced less NO_x pollution, both with and without the use of additives, than is common with conventional plant. The reason for the superior performance is uncertain. NO_x emission could not be correlated with SO_2 emission, although on some occasions a decrease in the SO_2 emission (due to feeding limestone or dolomite) was accompanied by an increase in the NO_x emission. It was concluded that more information was needed on the mechanism of NO_x formation before a contribution could be made towards reducing emission.

Emission of Alkalis and Chlorine

As expected, the low combustion temperatures in fluid-bed combustors resulted in low alkali emissions. The combustion gases from the pressure combustor contained about 2 ppm of Na; i.e., about one tenth of the lowest concentration reported for the gases from conventional plant. The concentration of K was less than 0.5 ppm. Higher emissions were measured when limestone was added to the pressurised combustor instead of dolomite (5 ppm of Na and 1.5 ppm of K) and from one of the non-pressurised combustors that was being operated at the higher bed temperature of 1560°F (6 ppm of Na and 3 ppm of K). As expected most of the chlorine of the coal was released into the combustion gases.

Emission of Particulates

Particulate matter elutriated from fluidised-bed combustors comprises 5 to 15 percent of the carbon and 80 to 100 percent of the ash and additive. By using primary and secondary cyclones having collection efficiencies of 90 percent at about 10 μm it was possible to collect 95 - 98 percent of this material to give dust emission of 0.2 - 0.6 gr/scf. Within this range the emission was approximately proportional to the feed rate of ash plus additive. Increasing the fluidising velocity increased elutriation from the bed, but because of more efficient cyclone operation with higher gas flow rates there was little effect on emission. Fines recycle in the 36-in. combustor increased the dust emission to 1.4 gr/scf. The pressurised combustor had an internal recirculation cyclone in addition to primary and secondary cyclones; dust emissions in the range 0.05-0.1 gr/scf were obtained.

Based on these results it is unlikely that there would be any problem in meeting projected statutory limitations on particulate emission.

Corrosion and Deposition

The addition of limestone or dolomite had no significant effect on corrosion or deposition of tubes in the bed or in the gas space under the range of operating conditions likely to be experienced in a commercial plant.

The amount of material settling on the turbine blade cascade at the outlet of the pressure combustor was slight and was judged to be unlikely to affect turbine performance. There were no signs of sintered deposits or erosion.

CONCLUSIONS

The main conclusions reached from the work are:

(1) With fluidised combustion and the addition of limestone (or dolomite) the

emission of sulphur oxides from coal burning power plant can readily be controlled to meet the very rigorous restrictions (100 ppm v/v SO_2) planned for certain densely populated areas in the U.S. For a power plant burning a 3 percent sulphur coal this would involve feeding sufficient additive to retain 95 percent of the sulphur. Under the best combinations of operating conditions about 1.8 times the stoichiometric quantity of additive would be required; for a 100-MW plant this would involve supplying 160 ton/day of limestone or 280 ton/day of dolomite. The less stringent restrictions that have been proposed for built-up areas (300 ppm), and for power stations generally in the U.S. (700 ppm), would require sulphur retentions of 85 percent and 67 percent respectively, for 3 percent sulphur coals. These limits can be met under a wider range of operating conditions and/or at less expense for additives.

(2) Emission of oxides of nitrogen from fluidised combustion systems can be expected to be at least 60 percent less than from conventional combustion systems but additional measures would be needed over and above those used for SO_2 reduction to meet the very stringent restrictions envisaged for the latter part of the century (i.e., 100-200 ppm).

(3) The particulates emitted from fluidised-bed systems are unlikely to cause problems in meeting current or possible future restrictions.

(4) The use of limestone/dolomite additive to restrict sulphur emission is unlikely to affect adversely the exemplary behaviour of the fluid-bed combustion system from the point of view of (a) fouling and corrosion of tubes immersed in the bed and (b) deposition or erosion of turbine blade materials exposed to the combustion gases.

In terms of sulphur retention the most important variable is the Ca/S mole ratio. The most stringent requirement for SO_2 emissions yet proposed can readily be met if sufficient calcium is present in the bed. If economic factors require it, the usage of limestone/dolomite can be minimised, by reducing the design fluidising velocity. This will make the boiler bigger and hence more expensive.

In terms of boiler operation and control the most important variable is bed temperature. It has been shown that under some conditions the efficiency of sulphur retention is very sensitive to bed temperature; for ease of boiler start-up and flexibility during load following it is useful to be able to let the bed temperature vary through the maximum allowable range. For American coals this range is probably from about 1460 to 1800°F.

ACKNOWLEDGMENTS

The author wishes to thank the National Coal Board for permission to publish this paper. Any views expressed are his own and not necessarily those of the Board.

This paper is no more than a brief summary of research work which was fully written up in a Main Report and nine Appendices involving the efforts of more than 20 people.

The author, therefore, gratefully acknowledges the efforts of all the following:

The NCB Contract Manager was D. H. Broadbent, assisted by S. J. Wright. The research programme was directed by A.D. Dainton (CRE) and H.R. Hoy (BCURA). The project co-ordinator was D.J. Loveridge. The pilot plant experimental work at CRE was administered by J. McLaren. The following personnel were involved in the tasks into which the programme was divided.

Task I	The project leaders were D. C. Davidson and D. F. Williams; A. A. Randell was responsible for operation of the plant; D. G. Cox and J. Highley carried out the data processing and assessment of results.
Task II	The project leader was A. G. Roberts; D. M. Wilkins was responsible for operation of the plant; J. E. Stantan carried out the data processing and assessment of results.
Task III	The project leader was D. J. Loveridge; M. H. Barker was responsible for operation of the plant; D. M. Wilkins carried out the final data processing and assessment of results.
Task IV	The project leader was M. J. Cooke; B. J. Bowles was responsible for operation of the plant; E. A. Rogers carried out the corrosion studies.
Task V	The project leader was D. C. Davidson; A. W. Smale was responsible for operation of the plant; D. G. Cox, J. Highley and J. Holder carried out the data processing and assessment of results.
Task VI	The project leader was D. W. Gill; he was assisted by F. V. Bethell and B. B. Morgan.
Task VII	The project leader was D. C. Davidson; the experimental work was carried out by R. F. Littlejohn.
Task VIII	The project leader was D. H. T. Spencer; he was assisted by A. A. Herod and B. A. Napier. Additional work to obtain data for the mathematical model was carried out by F. V. Bethell and G. McDonald.

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The OAP representative in the U. K. was E. L. Carls. His valuable contribution both in the experimental work and in the preparation of the progress reports and the final report is acknowledged.

GLOSSARY OF TERMS

Absorption ratio: SO_2 reduction divided by 100 minus SO_2 reduction.

Ca/S mole ratio: moles of calcium in additive divided by moles of sulphur in coal.

Excess air: air input minus stoichiometric air for coal input divided by stoichiometric air for coal input, times 100 percent.

Fluidising velocity: volume flow rate of gas at combustion temperature and pressure divided by cross section of combustor (neglecting tubes).

SO_2 reduction: SO_2 emission without additive minus SO_2 emission with additive divided by SO_2 emission without additive, times 100 percent.

Sulphur retention: sulphur in coal minus sulphur in gas divided by sulphur in coal, times 100 percent.

Unburnt carbon loss: unburnt solid carbon divided by carbon in coal input, times 100 percent.

Utilisation of additive: moles of sulphur retained by additive divided by moles of calcium in additive, times 100 percent.

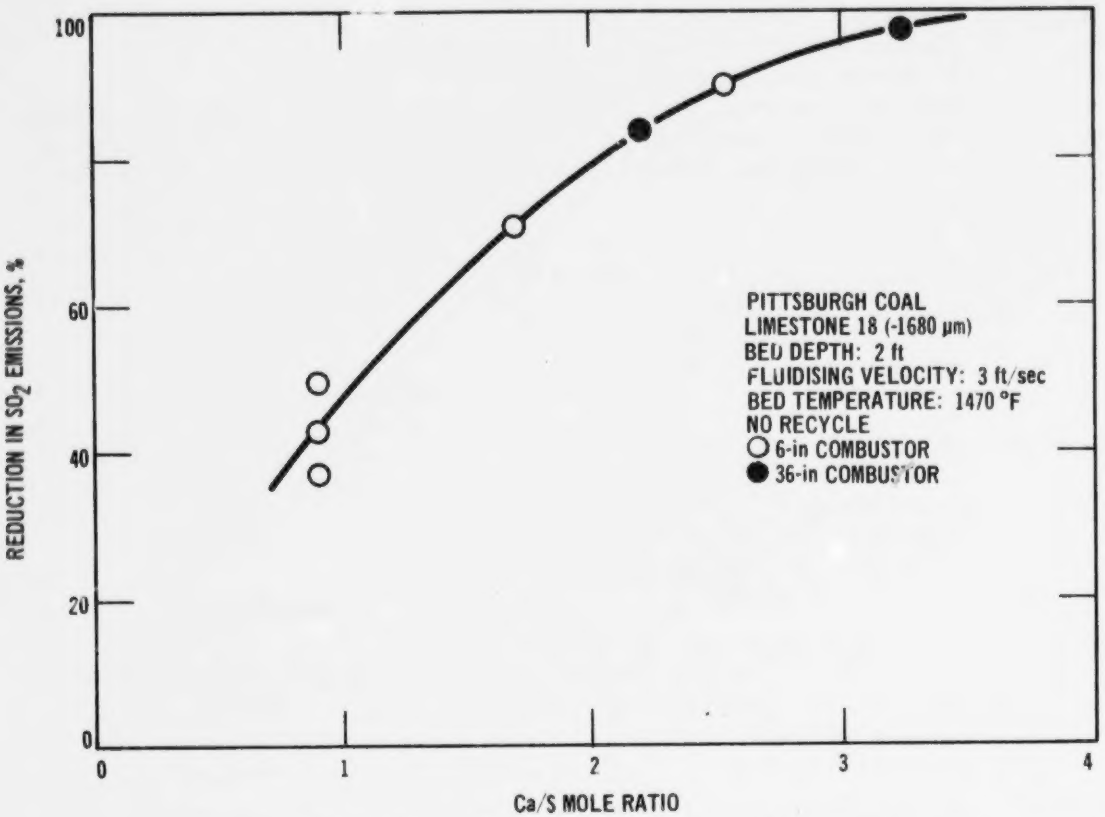


Figure 1. Comparison of SO_2 reduction in 6-in. and 36-in. combustors.

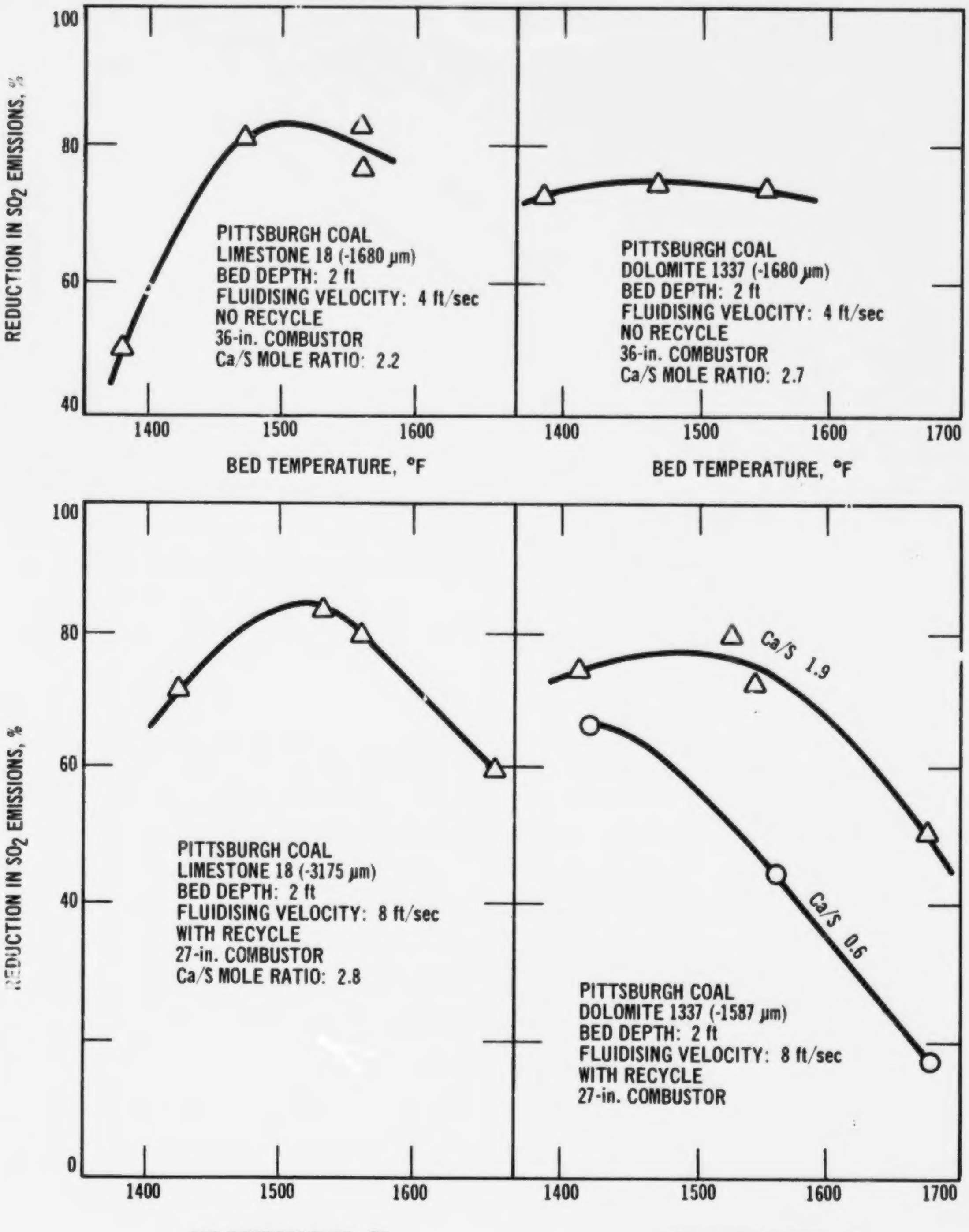
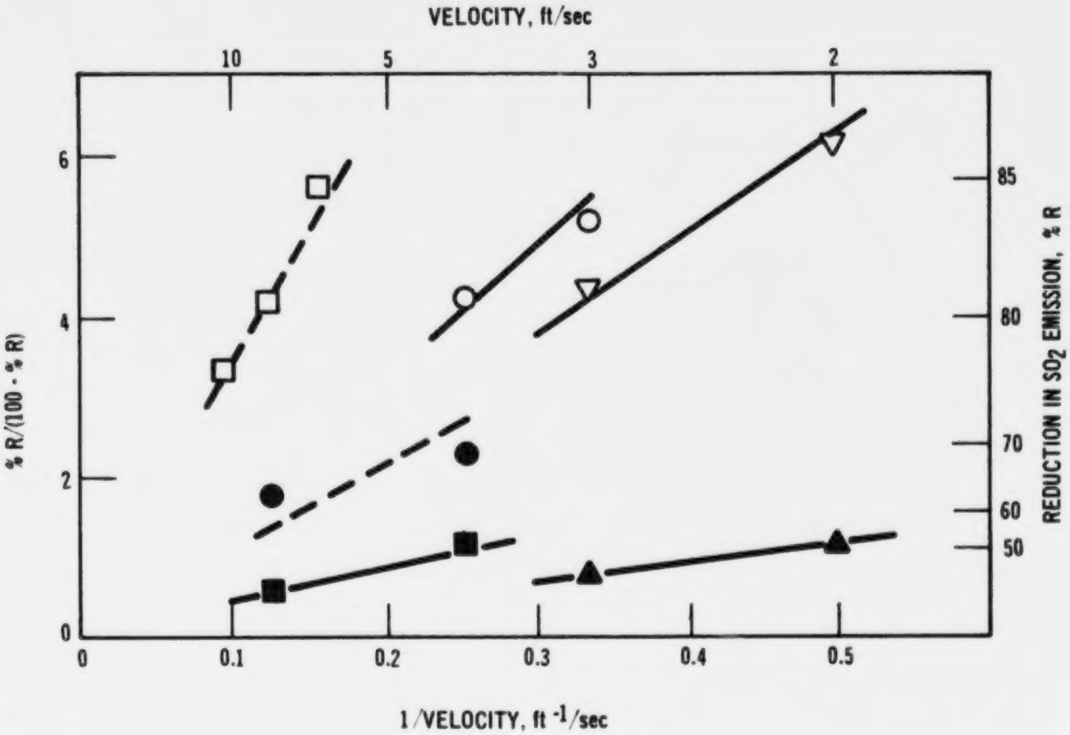


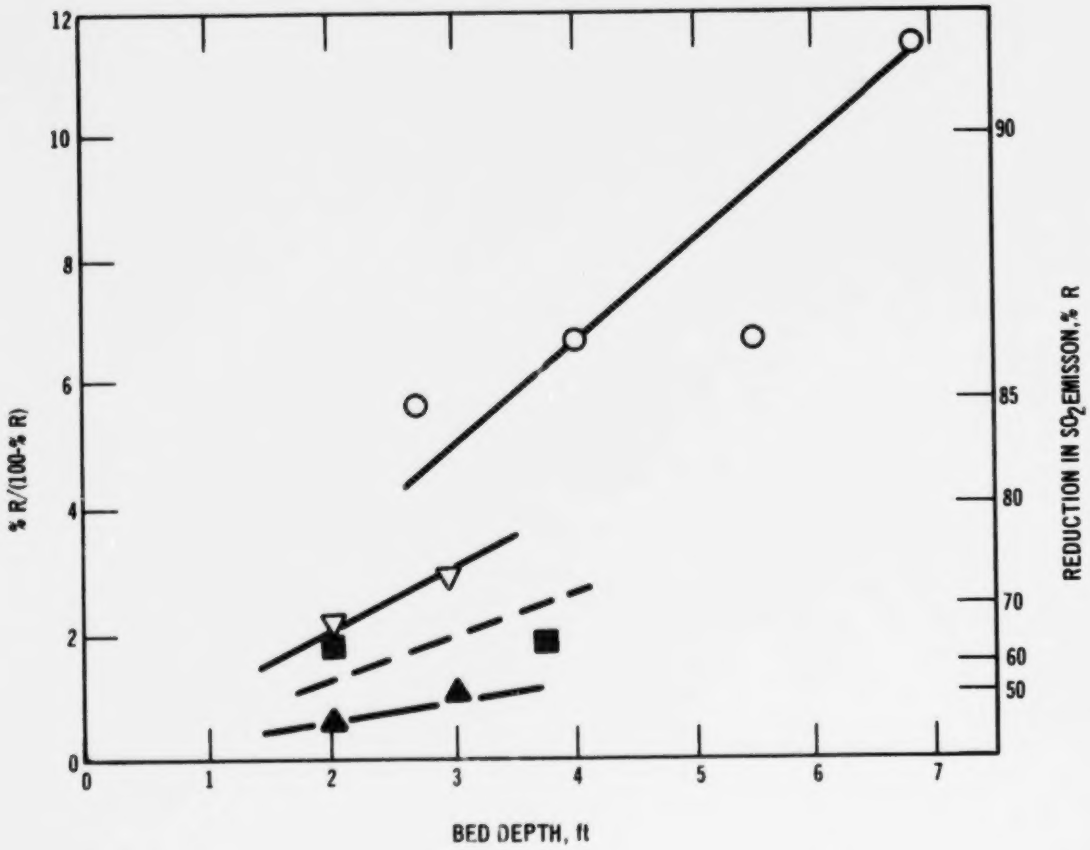
Figure 2. Effect of bed temperature on SO_2 reduction.



SYMBOL	COAL	ADDITIVE	SIZE, µm	BED DEPTH, ft	BED TEMP., °F	Ca/S MOLE RATIO
■	PITTSBURGH	18	3175	2	1560	1.7
○	"	"	1680	2	1470	2.2
▲	WELBECK	U.K. L'STONE	"	2	"	0.8
▽	"	"	"	2	"	1.8
●	"	"	3175	2	1560	2.8
□	PITTSBURGH	18	"	2	"	2.7

36-, 27-, AND 6- in. COMBUSTORS

Figure 3. Empirical relation between SO_2 reduction and fluidising velocity.



SYMBOL	COAL	ADDITIVE	SIZE, μm	VELOCITY, ft/sec	BED TEMP., ° F	Ca/S MOLE RATIO
■	WELBECK	U.K. L'STONE	3175	8	1560	2.8
○	PITTSBURGH	1337	"	"	"	5.0
▲	ILLINOIS	1359	1680	3	1470	1.1
▽	"	"	"	"	"	2.2

36- AND 6-in. COMBUSTORS

Figure 4. Empirical relation between SO_2 reduction and bed depth.

5. SELECTIVE EXTRACTION OF CLINKER AT THE BOTTOM OF A DEEP SELF-AGGLOMERATING FLUIDIZED BED

A. A. GODEL

Societe Anonyme Activit

In our former Conferences, very few communications have dealt with self-agglomerating fluidized beds. To my knowledge, the only case of the subject which has been studied — at least in the field of industrial achievements — was the use of agglomerating fluidized beds for coal combustion via the "Ignifluid Process" which has been the subject of several reports given here: in 1968, by myself on behalf of the Activit Company, and in 1970, by Mr. Svoboda, Manager of the Babcock-Atlantique Company, and by Mr. Demmy, Vice-President of the U.G.I. Corporation.

Professor Squires has honored me by crediting me with having brought to light the process in which self-agglomeration in a heavily turbulent fluidized bed results from preferential bonding of slag particles when they reach their sintering temperature (about 1100°C for most coal ash); on the contrary, when slag particles—even adherent ones—encounter coal particles, there is little chance of their agglomeration. This phenomenon stems from laws governing the probability of encounter of particles.

The result of our study, quite fortunately, is the possibility of forming slag agglomerates in a state of quasi-purity. Slag agglomerates fall to the bottom of the bed when they acquire sufficient weight and must be eliminated promptly in order to avoid blocking fluidization. Extracting ash by slag agglomeration is doubly interesting, for it also permits reinjecting the dust carried over into the fluidized bed since it has no excess of cinders.

In most cases, this leads us to prefer using agglomerating fluidized beds which require no temperature limit.

As I stated here in 1968, the Ignifluid combustion process achieves extraction quite simply by using an inclined upward-moving fluidization grate. This grate supports the fluidized bed with clinker deposited at the bottom; it passes through the surface of the bed, operating thereby like a clinker extractor.

Such grates are relatively narrow and may be easily placed under rectangular boilers. This position has enabled their successful industrial development in equipping boilers of various steam output for more than 16 years by the Babcock-Atlantique Company (exclusive license-holder for the process). A 60-MW Ignifluid power plant comprising two boilers has been in successful operation for the past three years. Equipping larger boilers raises no problems; several projects have been designed with such equipment and exported from France since the use of coal is constantly decreasing in Europe.

Having stated this, I should like to discuss the essential reason for my communication—a new process for extracting slag at the base of a deep fluidized bed. This new process is quite different from the former in both the means used and the aims envisaged, although both the Ignifluid Process and the new one operate in self-agglomerating fluidized beds.

Our goal in the new process is principally to achieve certain chemical reactions in deep fluidized beds, sometimes under pressure; e.g., the processing of mineral ore, the processing of chalky marl for producing cement clinker, etc.

It should be noted that the new process may be adapted to the use of beds for fuel gasification in conjunction with appropriate complementary treatments, such as desulphurization and, in certain cases, catalytic reactions.

In my invention, I have been guided by the obvious principle that it is easier to sort slag in a shallow bed than in a deep one. I have therefore combined the use of two fluidized beds of different depths, profiting from the communicating vessel principle or the "diving bell" principle, as you will see in Figure 1. (The figures are produced simply to give a theoretical explanation of the process.)

Figure 1 shows a horizontal grate supporting a fluidized bed (A) of a certain depth and containing slag at its base. I shall call this bed the "principal fluidized bed." On the right, communicating with the first one by opening (O), is a shallow fluidized bed (B), also containing slag at its base, which I shall call the "auxiliary fluidized bed."

As you will notice, the auxiliary fluidized bed is contained in a closed space (C), constituting what I shall call a "fluidization cell."

Since gas escaping from the auxiliary fluidized bed finds no outlet to the right, it escapes on the left through opening (O) which communicates with the principal fluidized bed.

Under these conditions, the auxiliary fluidized bed is in hydrostatic equilibrium with the principal fluidized bed; i.e., the pressure is the same at corresponding levels.

Theoretically, the surface of the auxiliary fluidized bed should thus be horizontal; but the fluidization gas leaving this bed and flowing laterally through its surface toward one end causes the surface to assume a concave shape, dropping away considerably toward the opposite end, as shown.

Figure 2 shows the same theoretical layout, but an air intake is provided at the end of the fluidization cell by opening (d) in order to increase the flow escaping through (O) toward the principal fluidized bed (A). The result is that the surface of the auxiliary fluidized bed (B) drops away much more quickly than shown in Figure 1, until it lays bare the slag lying on the bottom of the auxiliary fluidized bed. Perfect separation is thus achieved between slag particles and the fluidized matters.

To practically implement the new process, we use a cylindrical or cylindro-conical reactor, fitted at its base with a circular fluidization grate which supports a principal fluidized bed and a very shallow auxiliary fluidized bed, the latter being contained in a small fluidization cell immediately underlying the principal fluidized bed.

The grate and the fluidization cell are in relative motion to one another; the fluidization cell is wide open at the base to permit communication between the two fluidized beds, which are in hydrostatic equilibrium as I mentioned previously.

The direction of the motion of the grate in relation to that of the cell is such that the slag deposited on the grate penetrates into the cell, where slag particles are decanted, extracted, and sorted outside the principal fluidized bed and finally poured into an ash pit in a state of quasi-purity.

These principles offer the possibility for a variety of embodiments, of which I shall mention only two.

The isometric projection in Figure 3 represents a cylindrical reactor (a) with a horizontal grate (b) revolving around its vertical axis (x-y). The fluidization cell (c) is fixed and provided with an outlet (d) toward an outside ash pit, not shown.

The reacting and fluidizing gas is injected by pipe (e); gas resulting from the reaction is evacuated by pipe (f) after having passed through a cyclone (g), which assures collection and reinjection of fine dust into the fluidized bed. Pipe (h) supplies the reactor with granulated solid material to be treated.

The grate revolves counter-clockwise, which causes the slag particles deposited on the grate to penetrate into the cell via opening (O). The size of this opening depends on the size of the apparatus, and may reach 30 or 40 cm (1 ft) for a large apparatus in order to avoid blocking by slag.

For a better understanding of the manner in which the apparatus shown in Figure 3 operates, I should like to ask you to refer to Figure 4 which shows an isometric projection of the fluidization cell at a larger scale. This cell is provided with vertical side-walls (a-b-c-d) topped by an upper wall forming a sealed cover.

Inside the cell, the grate supports the auxiliary fluidized bed which communicates with the principal fluidized bed via opening (O) situated at the base of wall (a), as mentioned previously.

The shape of the various vertical cell walls must be adapted to their specific functions, as follows:

1. Wall (a) is open at the base at (O) to establish the communication between the two fluidized beds and to provide a passage for the slag.

2. Wall (b) is identical to the reactor cylinder wall; it contains an opening (g) giving access to an ash-pit placed outside the reactor (not shown).

3. Wall (c) is preferably laid out in the shape of a logarithmic spiral, with its convex face turned toward the outside of the cell to force slag particles brought in by the revolving motion of the grate toward opening (g). This arrangement profits from the very special fact that this curve has, at all points, a constant angle in relation to the incident direction of slag brought in by the grate (for this purpose, an angle of 30° is very favorable). Another essential function of wall (c) is to prevent the principal fluidized bed from entering the cell on this side and flooding it.

4. Wall (d) serves as a simple connecting element between walls (a) and (c).

This is only a schematic outline of the cell for, in fact, the cover of the cell will be preferably in the shape of a peaked roof, following the angle of repose or natural slope of products being processed.

The second example which I should like to illustrate is shown in Figure 5. Here you see a cylindro-conical fluidization reactor (a) containing in the center an ash pit surrounded by a fixed annular horizontal grate (b).

The fluidization cell (c) revolves around its vertical axis (x'-y') and has an evacuation toward the inside ash pit situated at (d), which is also rotating.

Reacting gas is injected into the reactor at (e); gas resulting from the reaction is evacuated by pipe (f) after having passed through a group of two cyclones (g) mounted in series; the latter collect and reinject flue dust into the fluidized bed.

The reactor is supplied with granulated solid material in this example through pipe (h) so that solid material circulates by counter-current flow with relation to gases in interest of heat recovery.

In this second type of installation the grate is fixed; the relative motion between grate and cell results from the cell's own clockwise motion to assure the penetration of slag into the cell by opening (O').

The arrangement of the fluidization cell (c) is based on the same principle as in the preceding example (Figure 4). However, wall (c) in Figure 6 is curved in the shape of a logarithmic spiral with the concave face turned toward the inside in order to force slag particles toward the inside ash pit.

You will note that wall (a') is wide open at the base in (O') to permit inserting the fluidized bed and the slag particles. Vertical wall (b') is circular. Vertical wall (c') is laid out in the shape of a logarithmic spiral with concave part turned inwards, as already mentioned. As with the preceding cell, it is closed by a sealed cover.

From the point of view of the mechanical fabrication of the various parts, I feel that it is unnecessary to enter into details concerning them, except to mention that the cell must be cooled by circulating water or by vaporizing water.

Circular fluidizing grates are of the ordinary type and may consist of refractory cast-iron links supported by an appropriate frame.

Startup is realized by a gas or fuel oil burner (not shown) placed in the upper part of the reactor. fired on top of the bed and is then static. Fluidizing gas (in fact, air) must be injected only when the temperature of the bed has reached a sufficiently high level.

For example, if the fluidized bed consists of coal, processing capacity may be estimated at about 2 to 3 tons/hr of coal gasified/sq meter of fluidizing grate.

The resulting processing capacity could be for an 8-meter diameter reactor, for example 150 to 200 tons of coal gasified/hr at atmospheric pressure. This capacity could be considerably increased to reach an equivalent power production of 1000 MW if pressure gasification is used.

Naturally, reactors designed according to this new technique may be perfected in various manners, particularly in view of obtaining high temperatures which are necessary for producing cement, by heating the fluid bed with coal fines or fuel oil and recovering heat carried off by gases with care. For this purpose, one might use a series of superimposed cyclones in which processed solid material circulates against the current of reaction gases (Figure 5).

In addition, residual heat of reaction gases may be recovered by diverting these gases to a heat exchanger.

Finally, for gasification or for other chemical reactions at very high temperatures (500 to 1000°C), reacting gases may be injected by nozzles (Figure 5, "j") into the truncated part of the reactor. This injection, if sufficiently great, may give rise to a dilute fluidized bed, which obviously must be stabilized at the top by cyclones adapted for this purpose. This latter use of dilute fluidized bed permits, as it is well known, a considerable increase in unit production.

From all that has been stated, I feel that one may conclude that the process offers the advantage of an exceptionally simple achievement with a consequent low cost, given its production capacity.

This process has undergone successful cold experiments on different types of 1:20 scale models. The development is so recent that no industrial nor semi-industrial testing has been carried out but a pilot unit is now in the course of realisation for the gasification of 250 to 750 kWhr of coal. So far, no major difficulties have appeared.

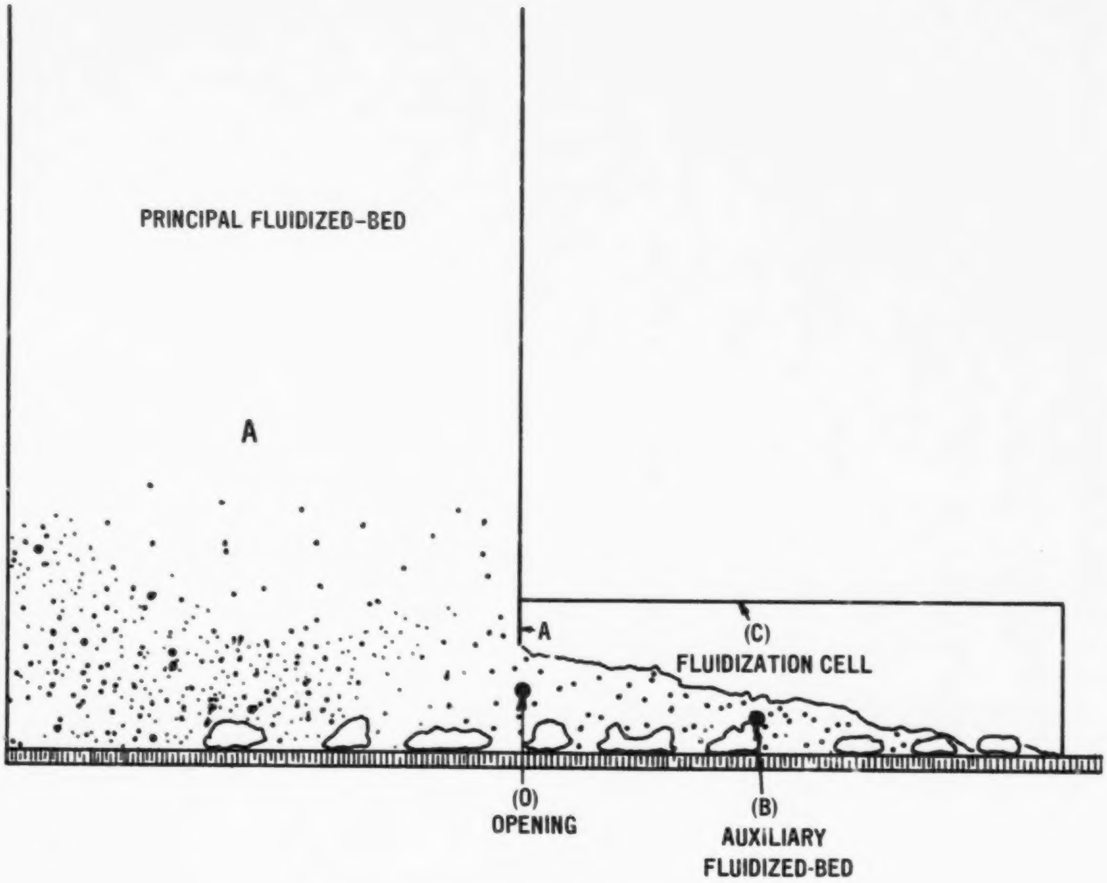


Figure 1. Fluidized-beds of different depths.

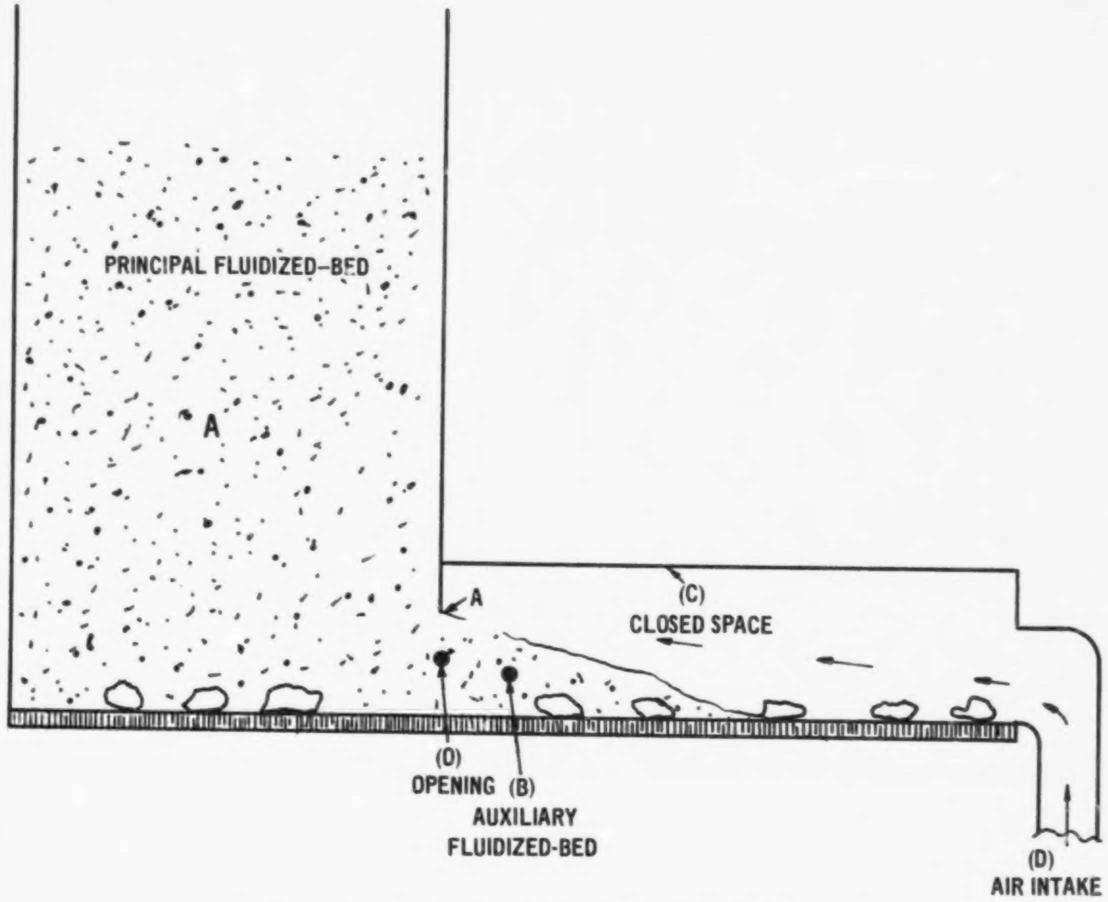


Figure 2. Fluidized-beds of different depths with air intake.

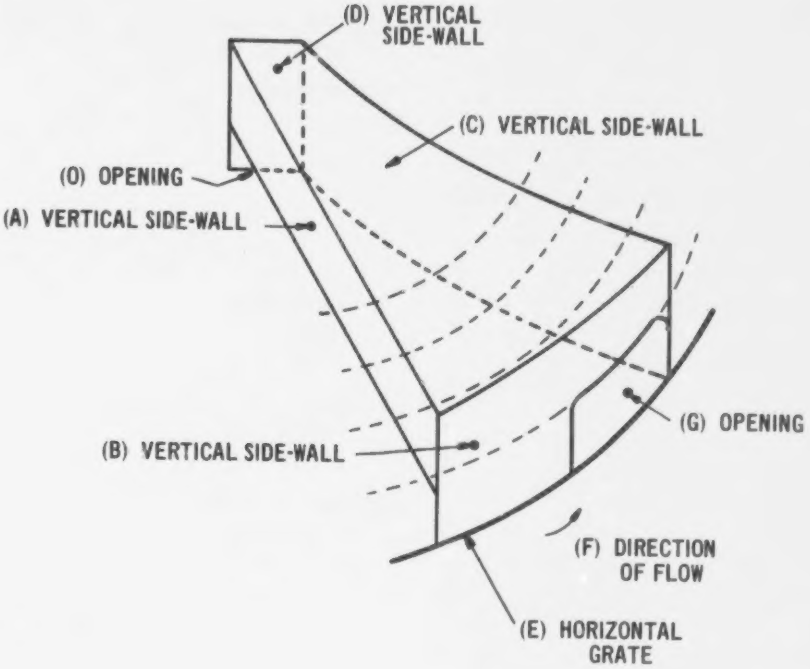


Figure 4. Detail of isometric projection of the fluidization cell.

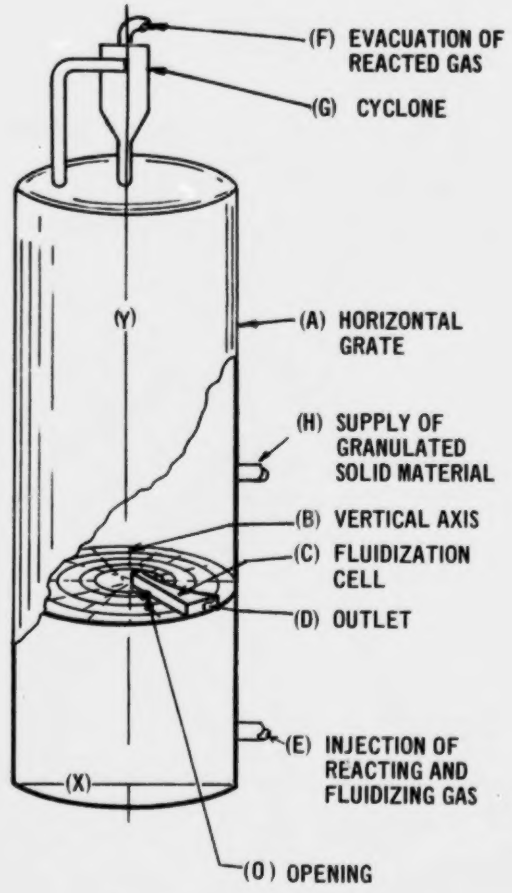

(F) EVACUATION OF REACTED GAS
(G) CYCLONE
(Y)
(A) HORIZONTAL GRATE
(H) SUPPLY OF GRANULATED SOLID MATERIAL
(B) VERTICAL AXIS
(C) FLUIDIZATION CELL
(D) OUTLET
(E) INJECTION OF REACTING AND FLUIDIZING GAS
(X)
(O) OPENING

Figure 3. Cylindrical reactor.

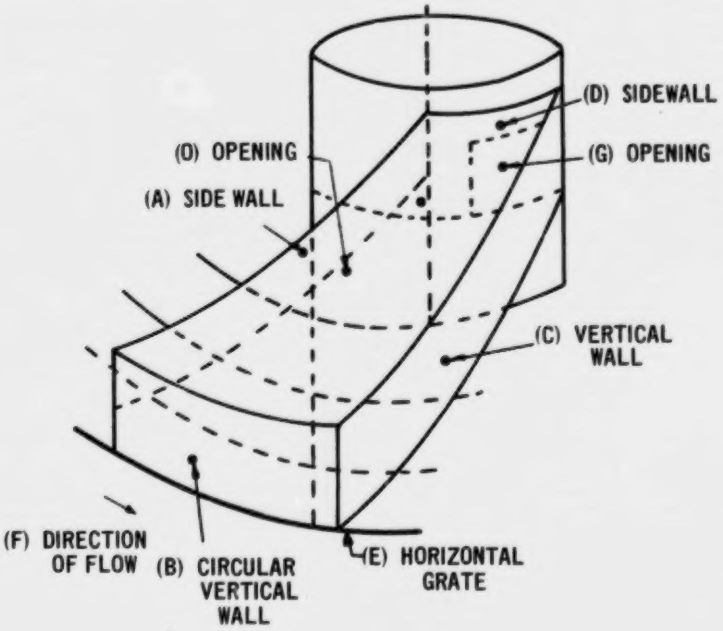


Figure 6. Detail of fluidization cell.

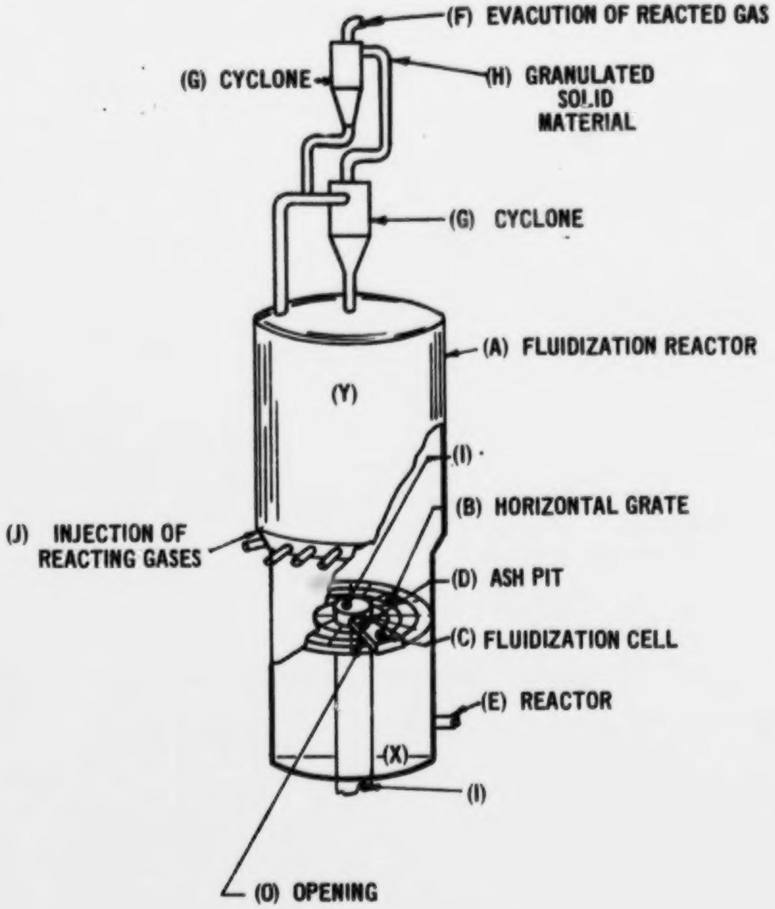


Figure 5. Cylindrical reactor.

6. KINETIC STUDIES RELATED TO THE USE OF LIMESTONE AND DOLOMITE AS SULFUR REMOVAL AGENTS IN FUEL PROCESSING

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ABSTRACT

A pressurized thermogravimetric analysis system adapted to handle corrosive gases has been used to obtain sulfur removal and regeneration data for limestone and dolomite. The kinetic feasibility of the desulfurization processes proposed for fluidized-bed gasification and combustion has been demonstrated at ten atmospheres. Calcium carbonate can be regenerated for gasification processes. Regeneration for the combustion system requires further study.

INTRODUCTION

Power generation through fluidized-bed fossil fuel processing, which uses limestone or dolomite in the bed to capture sulfur, has the potential to meet SO_2, NO_x and particulate pollution abatement goals at reduced energy cost. The application of fluidized beds to the gasification and combustion of oil and coal at elevated pressures, with combined cycle power generation, and to oil gasification at atmospheric pressure for retrofit on conventional plants, is being developed.[1] The efficacy of sulfur removal is a major criterion in evaluating these concepts.

Four aspects of the chemistry of sulfur removal will directly affect the usefulness of limestones or dolomites as traps for sulfur in fluidized-bed gasification and combustion of fossil fuels. The rate of sulfur removal from the bed gases during gasification or combustion and the capacity of the stones are primary concerns for all the proposed systems; regeneration of spent stone in an active form with concomitant sulfur recovery and disposal of waste stones which contain sulfur are of complementary importance.

The cycle of reactions in Figure 1 encompasses the reactions of concern. Table 1 lists the reactions and the conditions for the process options which are being assessed by Westinghouse.[1] Thermodynamic feasibility is necessary but insufficient for success of the proposed processes; a knowledge of the kinetics of the essential reactions under the process conditions can only be obtained by experiment. Despite atmospheric pressure data obtained by Pell et al.[4] on hydrogen sulfide reactions with dolomite, and various studies on sulfur dioxide "sorption" by limestones and dolomites[6,7,8] there is a need for primary kinetic data with which the behavior of the fluidized-bed desulfurization of fuels can be predicted and explained.

The objectives of the current program are to:

1. Establish which reactions occur,

2. Determine the reaction kinetics,
3. Determine stone utilization achievable,
4. Study the effect of regeneration on stone reactivity,
5. Probe reaction mechanisms,
6. Assess the influence of side-reactions,
7. Recommend optimal operating conditions.

In this paper we describe how a thermogravimetric analysis (TGA) system, designed to obtain the desired data, is being used to survey the chemistry of the processes, and how it is focusing attention on critical areas where successful use of the reactions requires further study.

Table 1. REACTIONS AND CONDITIONS OF WESTINGHOUSE PROCESS OPTIONS

Reaction	Operating conditions	Applicable fuel processing option
Sulfur removal		
1. $\frac{CaO}{CaCO_3} + SO_2 + 1/2\ O_2 \rightarrow CaSO_4 + \frac{-}{CO_2}$	1300°F$<T<$1750°F P: 1 to 20 atm	Combustion
2. $\frac{CaO}{CaCO_3} + H_2S \rightarrow CaS + H_2O + \frac{-}{CO_2}$	1300°F$<T<$1700°F P: 1 to 20 atm	Gasification
Stone regeneration		
3. $CaSO_4 + \frac{H_2}{CO} \rightarrow CaO + SO_2 + \frac{H_2O}{CO_2}$	$T>$2000°F	Combustion (low SO_2 concentration at elevated pressure)
4. $CaSO_4 + \frac{4\,H_2}{4\,CO} \rightarrow CaS + \frac{4\,H_2O}{4\,CO_2}$ (followed by reaction 5, 6, 7 or 8)	$T<$ 1600°F	Combustion (at elevated pressure)
5. $CaS + H_2O + CO_2 \rightarrow CaCO_3 + H_2S$	$T<$1500°F $P>$5 atm	Combustion/gasification (at elevated pressure)
6. $CaS + H_2O + CO_2 \rightarrow CaCO_3 + H_2S$	T~200°F atm	Combustion/gasification (not recommended due to low temperature & water purification)
7. $CaS + 3\,CO_2 \rightarrow CaO + 3\,CO + SO_2$	$T>$2000°F atm	Combustion/gasification (not recommended due to low SO_2 concentration, ~2%)
8. $CaS + 3/2\ O_2 \rightarrow CaO + SO_2$	$T>$1800°F	Combustion/gasification (low SO_2 concentration at pressure)
9. $CaSO_4 + C + H_2O \rightarrow CaCO_3 + CO_2 + H_2S$		Combustion (advanced concept)
10. $CaS + 2\ O_2 \rightarrow CaSO_4$	atm	All gasification

EQUIPMENT

The design conditions for both gasification and combustion (i.e., pressures of 10 to 30 atm, temperatures up to 1200°C, and the corrosive gas compositions of hydrogen sulfide and sulfur dioxide) define an area in which little or no kinetic investigations of gas-solid reactions have been reported. For this study, a duPont thermogravimetric balance was mounted inside a pressure shell, so that it could record continuously the weight changes of a solid suspended in a reacting gas stream of pre-selected composition at temperatures up to 1200°C. Figure 2 shows the apparatus; Figure 3 shows a closeup view of the balance; and Figure 4 is a diagram of the system. The corrosion prevention system is based on that described by Ruth.[5] Despite this precaution, the balance has a limited lifetime owing to corrosion; alternative designs are currently under investigation.

The advantages of a TGA are the ability to isolate chemical reactions for study, the relative ease with which the desired conditions can be attained and controlled, and the accuracy and rapidity with which reactions can be studied, from the point of view of the solid.

The chief disadvantage for our purposes lies in the fact that it is not a fluidized bed; translation of TGA results into likely fluid-bed behavior is difficult. A second disadvantage is the small size of the stone samples used (~ 10 mg)* which makes it occasionally difficult to use chemical analysis to assess the importance of minor competing reactions; there is little product for such physical characterizations as BET analysis.

Operation of the pressurized TGA was tested by studying limestone calcination.[9] When the operating conditions were suitably modified, kinetic results in reasonable agreement with studies at atmospheric pressure were obtained.

MATERIALS

Solids used in the experiments were limestone 1359, Tymochtee dolomite, dolomite 1337, sieve fractions (420 to 590 μm) except where stated. The gases, N_2, O_2, CO, CO_2, H_2S, and SO_2 were taken from commercial cylinders. Steam was generated in the manifold.

RESULTS

The strategy adopted was to examine each reaction in turn before proceeding to study cyclic behavior.

A preliminary survey of the principal reactions of Table 1 has been carried out; Table 2 summarizes the results.

Sulfur Removal

Combustion:

$$CaO + SO_2 + 1/2\ O_2 \rightarrow CaSO_4 \quad (1)$$

The reaction between sulfur dioxide and calcined dolomite 1337 in excess oxygen followed closely the results obtained by Borgwardt[7] in a differential bed reactor; e.g., rate at 10 percent Ca utilization:

TGA results (850°C) 2.40×10^{-5} [mole SO_3 • gram calcined dolomite^{-1}•sec^{-1}]

Borgwardt's data 2.10×10^{-5} [mole SO_3 • gram calcined dolomite^{-1}•sec^{-1}].

The rate of reaction became increasingly smaller at 40 percent Ca utilization at pressures of 1 atm, but at 10 atm rapid reaction continued beyond 70 percent reaction, as shown in Figure 5. The initial rate in the pressurized case is probably limited by the supply of sulfur dioxide to the solid in our particular reaction system.

Gasification:

$$\left(\frac{CaO}{CaCO_3}\right) + H_2S \rightarrow CaS + H_2O + (CO_2) \quad (2)$$

---

*10 mg is kinetically optimal for particle sizes being studied; balance can accept up to 1 gram samples.

Table 2. THERMOGRAVIMETRIC ANALYSIS DATA SUMMARY

Reaction	Original substrate 500-μm dia	Pressure, atm	Gas composition[a]	Temp., °C	Results	Conclusions	Future work
(1) $CaCO_3 + SO_2$ $+ 1/2\ O_2$ $\rightarrow CaSO_4 + CO_2$	Tymochtee Dolomite	1	0.25% SO_2 4% O_2	800-850	40% sulfation at 1 atm.		Higher temperature composition
	Dolomite 1337	10			90% sulfation at 10 atm	Makes once-through systems competitive	Investigate utilization at low SO_2 concentration
$CaO + SO_2$ $+ 1/2\ O_2$ $\rightarrow CaSO_4$	Tymochtee Dolomite Dolomite 1337	1 10	0.25% SO_2 4% O_2	800-850	Comparable to $CaCO_3$ results		
(2) $CaCO_3 + H_2S$ $\rightarrow CaS + H_2O + CO_2$	Tymochtee	1	15% H_2S 1.5% H_2S in N_2	600-845	90% sulfidation Some suppression at high H_2S concentrations	Attractive utilization and rate achievable	High pressure, fuel gas effect
	Limestone 1359	10	0.5% H_2S in N_2 CO_2	760	No sulfidation before normal calcination	Appears impractical under CO_2 pressures $>P_{equilibrium}$	Higher temperatures
(4) $CaSO_4 + 4\ CO$	Tymochtee Dolomite,	1	25% CO	750, 820	~90% yield	Gives poor substrate for R_xS	Investigate SO_2 loss and effect of initial sulfate porosity
$\rightarrow CaS + 4\ CO_2$	Dolomite 1337	10	$CO/CO_2 = 2/1$	850			
(5) $CaS + H_2O + CO_2$	Tymochtee Dolomite,	1	CO_2, H_2O + 25%	590-700	Depends on source of CaS	For gasification promising, for combustion, unproven	
$\rightarrow CaCO_3 + H_2S$	Dolomite 1337	10					
(10) $CaS + 2\ O_2$	Limestone 1359,	1	Air	400-950	7% yield	Appears impractical as impervious sulfate shell forms	Seek higher conversion
-- $CaSO_4$	Tymochtee Dolomite (4000 μm - 1000 μm)	1	Air	400-800	Up to 90% yield	Suitable for disposal at 800 °C	Check loss of SO_2

---

[a] The balance is nitrogen.

Sulfidation of dolomite at atmospheric pressure gave results in broad agreement with Pell's work[10] (Figure 6). Rapid rates and high yields are attained. Some suppression of reaction at high hydrogen sulfide concentrations and lower temperatures (700°C) was observed here, confirming Pell's finding.

In limestone sulfidation at pressure (10 atm) complete reaction was achievable when the stone was calcined; a shrinking shell model empirically describes the kinetics. Sectioning of partially reacted stone reveals an outer layer of calcium sulfide and an unreacted core of lime. No sulfidation was apparent before the normal calcination temperature.

REGENERATION

The two-stage regeneration of calcium carbonate from calcium sulfate has been studied. Reduction of calcium sulfate according to reaction

$$CaSO_4 + 4\,CO \rightarrow CaS + 4\,CO_2 \qquad (4)$$

was essentially complete at temperatures between 750 and 850°C, and 10 atm total pressure (Figure 7).

Regeneration of carbonate from the sulfide produced in reaction (3) above proved to be difficult. After 25 percent regeneration, the rate became extremely slow (Figure 8). By contrast, yields greater than 70 percent were obtained when the dolomite was directly sulfided with hydrogen sulfide (Figure 9). The latter regenerated carbonate was calcined, and its reactivity with sulfur dioxide was tested. It proved to react as rapidly as freshly calcined dolomite.

Unregenerated sulfide is concentrated at the core of the particle, as evidenced by sectioning the reacted stone.

STONE DISPOSAL

Spent limestone or dolomite from gasification processes contain calcium sulfide. This compound liberates hydrogen sulfide on contact with carbonated water, preventing its disposal in an untreated form. The problem is important for once-through and regenerative processes.

Conversion of the calcium sulfide to calcium sulfate before disposal, has been proposed.

$$CaS + 2\,O_2 \rightarrow CaSO_4 \qquad (10)$$

This reaction has been studied at atmospheric pressure using sulfided limestone, sulfided dolomites, and air as reactants.

Sulfided limestone 1359 cannot be oxidized to sulfate, probably due to formation of the same impervious layer observed when the stone is directly sulfated.[11] Surface reaction is observed but it rapidly decays. The reactivity can be renewed by lightly crushing the stone and repeating the reaction, which proceeds to an additional degree of oxidation about equal to the first stage. At high temperatures (900°C), about 1 percent reaction occurs extremely rapidly, followed by complete cessation of reaction. The exothermic nature of the reaction may have formed an impervious dead-burned lime layer on the solid surface.

Sulfided dolomite may be oxidized to calcium sulfate. At low temperatures (550°C) an initially rapid rate of reaction falls off after 14 percent of the sulfur has been oxidized within eight minutes. By contrast, 92 percent of the sulfide is oxidized within three minutes at a nominal temperature of 800°C. The stone temperature may be higher (Figure 10).

MODELS

The development of kinetic models which encompass the data, with a view to making predictions of the course of reaction in fluidized beds, is one of the goals of our investigation. Empirically, limestone sulfidation fits a contracting sphere model, in agreement with the physical form of sulfided stones. Reduction of sulfated dolomite by carbon monoxide is apparently first-order in sulfate. However, sufficiently detailed variation of the controlling parameters has not yet been sufficiently studied to permit predictions of reaction behavior over the wide range of parameters applicable to the proposed processes.

CONCLUSIONS

The TGA has provided information on the rates of reaction and the degree of utilization which can be achieved by the proposed sulfur removal, stone regeneration, and solid disposal processes.

Removal of sulfur dioxide and hydrogen sulfide: high stone utilization (>70 percent) has been achieved within 30 minutes.

Regeneration of calcium sulfate by the two-step, low temperature process: calcium sulfate can be reduced to calcium sulfide (> 90 percent). Calcium carbonate has not been successfully regenerated from the sulfide — <30 percent regeneration at practical temperatures.

Regeneration of calcium sulfide: calcium carbonate can be regenerated from calcium sulfide produced from H_2S—70 percent regeneration in 15 minutes. minutes.

Stone disposal: calcium sulfide has been oxided (90 percent) to calcium sulfate using dolomite.

These results are for a limited range of operating conditions. Further work is required to assess these reactions over the full range of operating conditions projected for the processes.

Future work is planned to:

1. Assess the low-temperature $CaSO_4$ regeneration process.
2. Study the one-step, high-temperature $CaSO_4$ regeneration process and other alternative processes.
3. Assess the activity of regenerated stone as a function of the number of sulfur removal/regeneration cycles.
4. Further study sulfation of stones for disposal.
5. Translate the kinetic data to fluidized-bed systems with the aid of experiments on a 2-in. hot-model fluid bed and pilot plant experiments conducted by Westinghouse and others.
6. Understand reaction mechanisms.

ACKNOWLEDGEMENTS

We thank Dr. D. H. Archer for guidance and support. We also acknowledge the technical assistance of Drs. F. P. Byrne and C. R. Wolfe of the Westinghouse Analytical Chemistry Department.

REFERENCES

1. Archer, D.H., et al. Evaluation of the Fluidized Bed Combustion Process. Summary Report. Westinghouse Research Laboratories, Pittsburgh, Pa. Prepared for the Environmental Protection Agency, Research Triangle Park, N.C. under Contract Number CPA 70-9. November 1971.

2. Keairns, D.L. Fluidized Bed Gasification and Combustion for Power Generation. In: Proceedings Frontiers of Power Technology, Oklahoma State University, Stillwater, Oklahoma, October 1972.

3. Archer, D.H. Fuel Processing Tailored to Environmental Needs. (Presented at American Chemical Society National Meeting. September 1972.)

4. Pell, M., R.A. Graff, and A.M. Squires. (Presented at meeting of American Institute of Chemical Engineers. Chicago. December 1970.)

5. Ruth, L.A., A.M. Squires, and R.A. Graff. (Presented at American Chemical Society Meeting, Los Angeles. March 1971.)

6. Coutant, R.W., J.S. McNulty, R.E. Barrett, G.G. Carson, R. Fischer, and E.H. Lougher. Summary Report. Prepared For National Air Pollution Control Administration, Cincinnati, Ohio, under contract Number PH-86-67-115, 1968.

7. Borgwardt, R.H. Environ. Sci. Technol. 4(1):59, 1970.

8. Borgwardt, R.H. and R.S. Harvey. Environ. Sci. Technol. 6(4):350, 1972.

9. O'Neill, E.P., W.F. Kittle, C.R. Wolfe, and L.M. Handman. Unpublished results.

10. Pell, M. Ph.D. Thesis, City University of New York. 1970.

11. Davidson, D.C. and J. Highley. The National Coal Board, London, England. Final Report to the Environmental Protection Agency, Research Triangle Park, N.C. Appendix 8, September 1971.

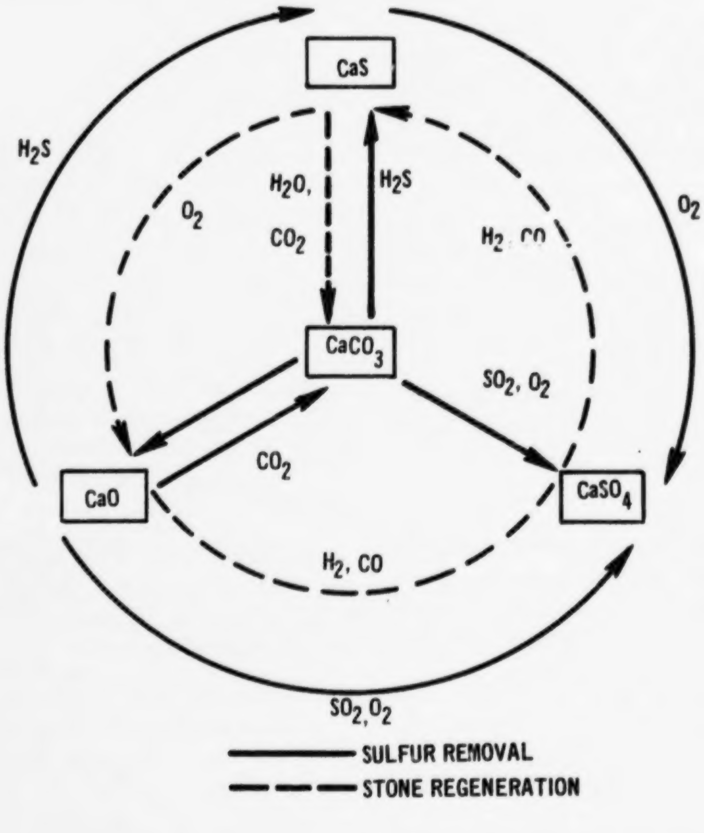

Figure 1. The calcium carbonate/sulfur cycle basic reactions.

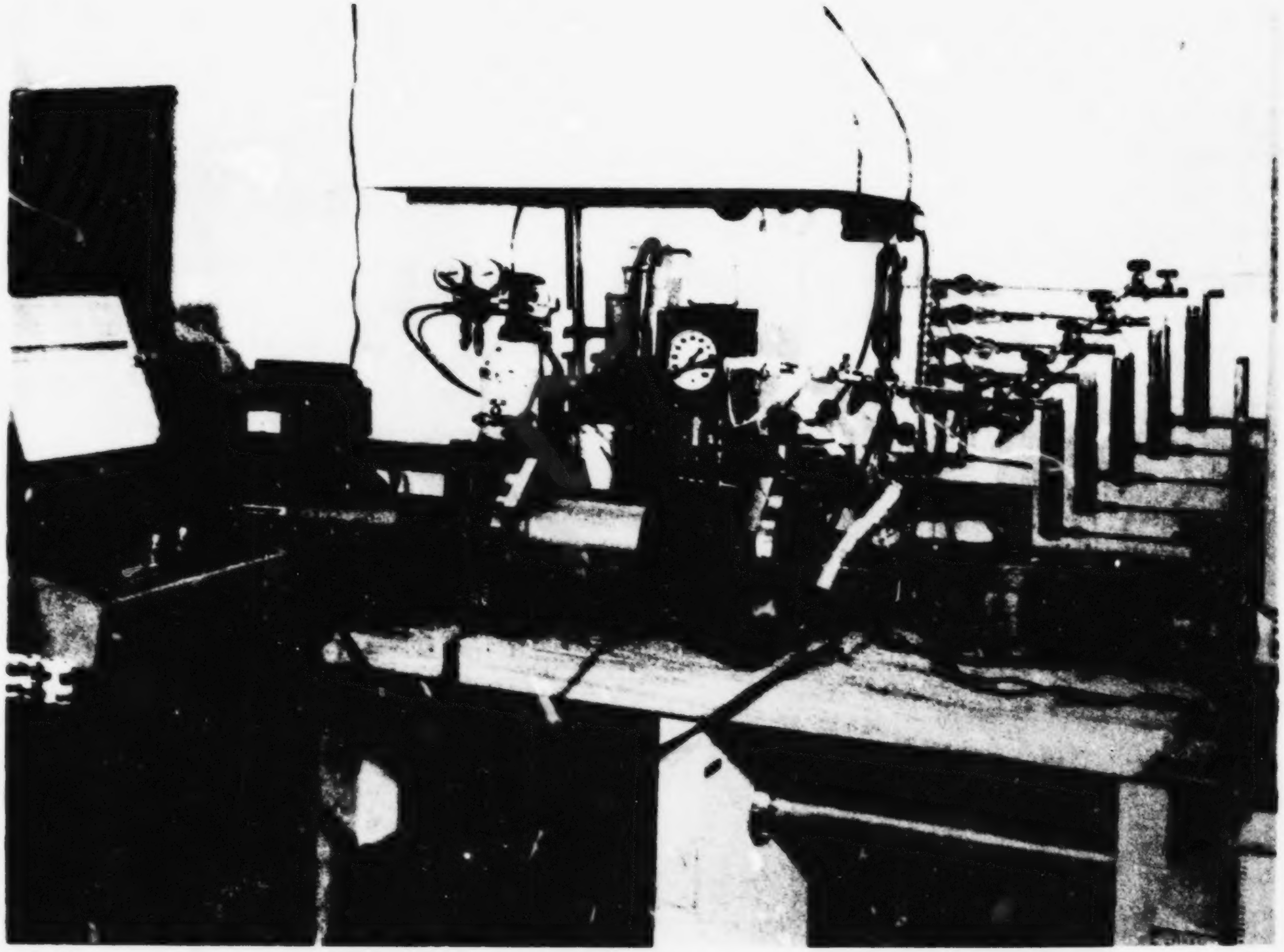

Figure 2. Thermogravimetric analyzer for high temperature and pressure reaction studies on limestone and char.

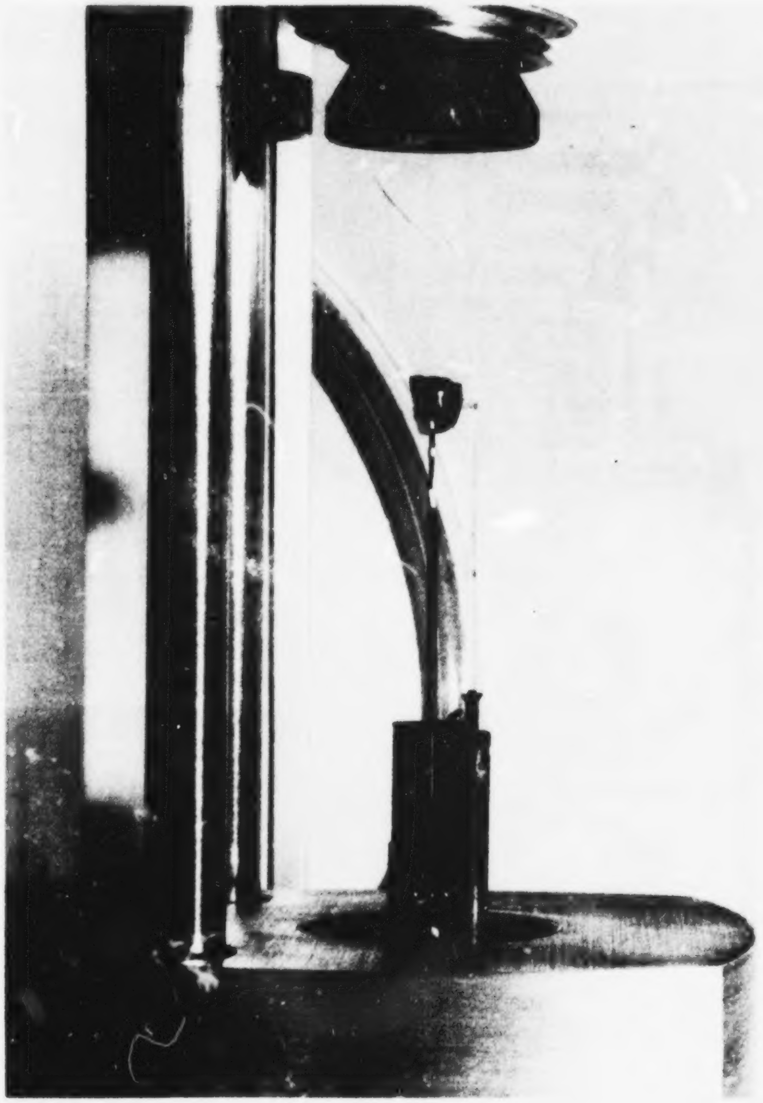

Figure 3. The duPont 950 thermogravimetric balance.

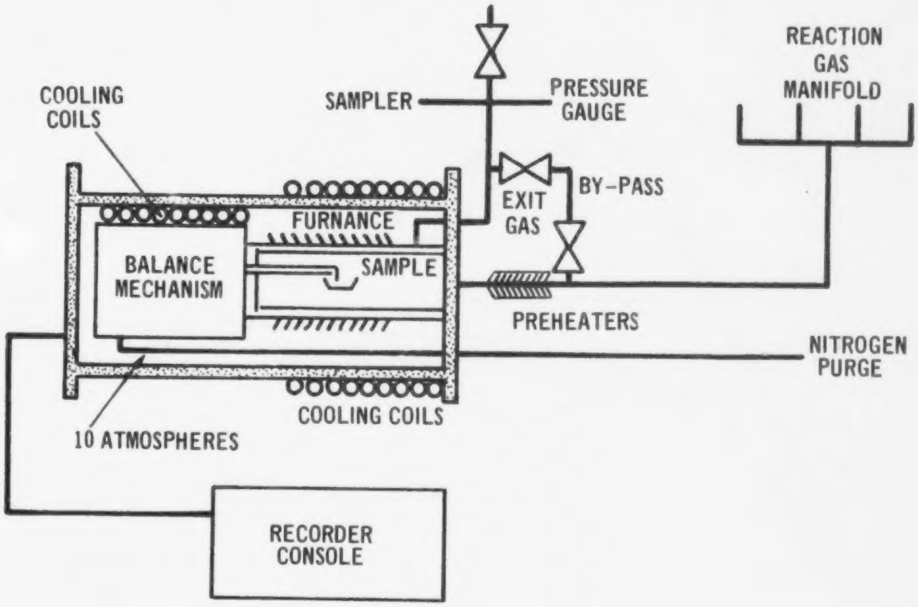

Figure 4. Diagram of the TG system.

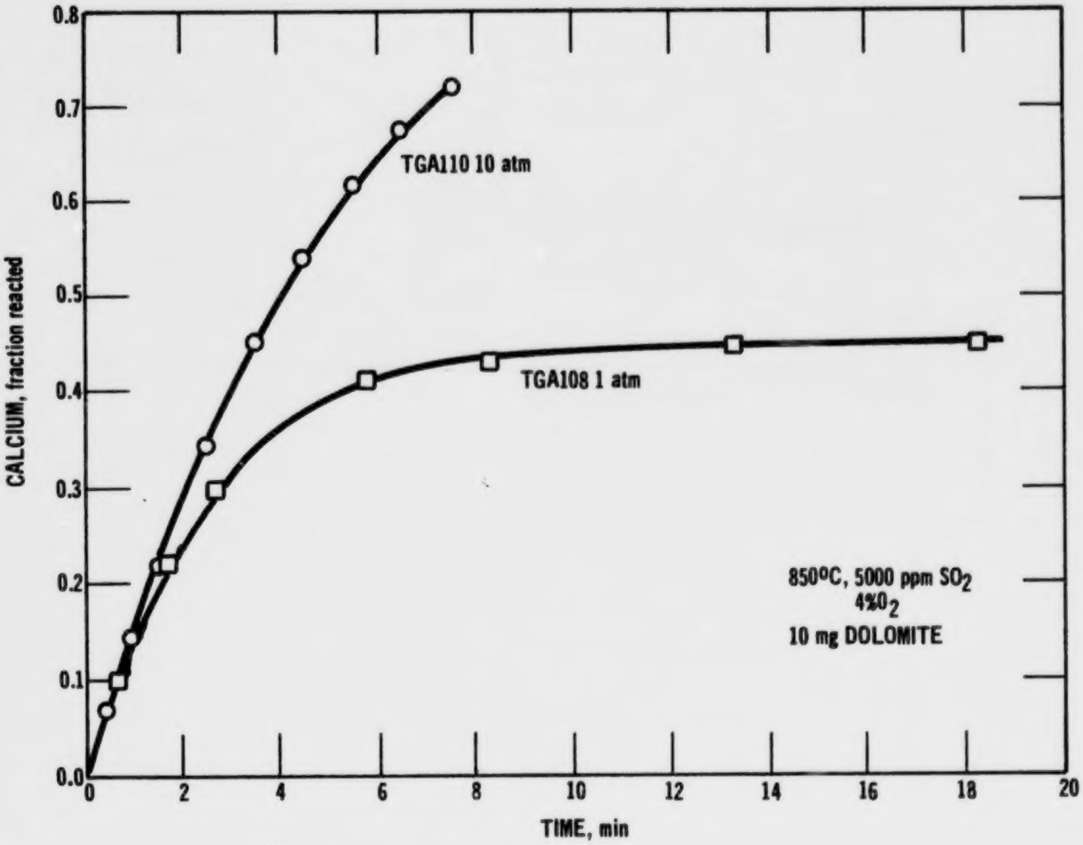

Figure 5. SO_2 reaction with calcined dolomite 1337.

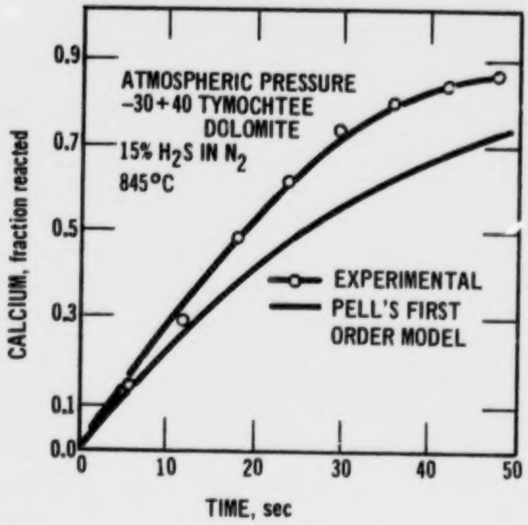

Figure 6. Sulfidation of calcined dolomite.

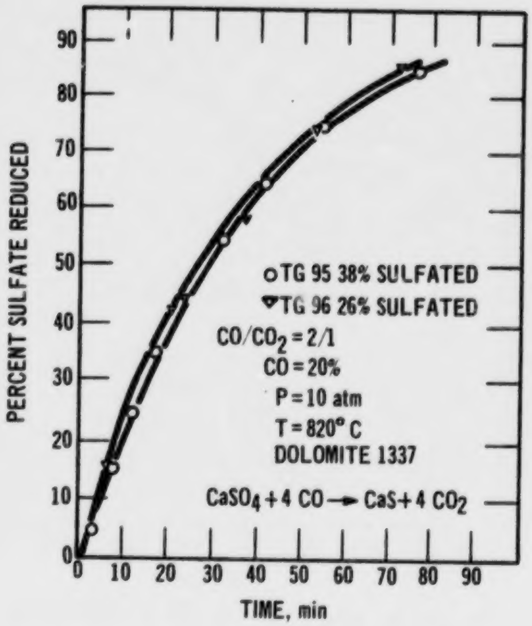

Figure 7. Reduction of sulfated dolomite.

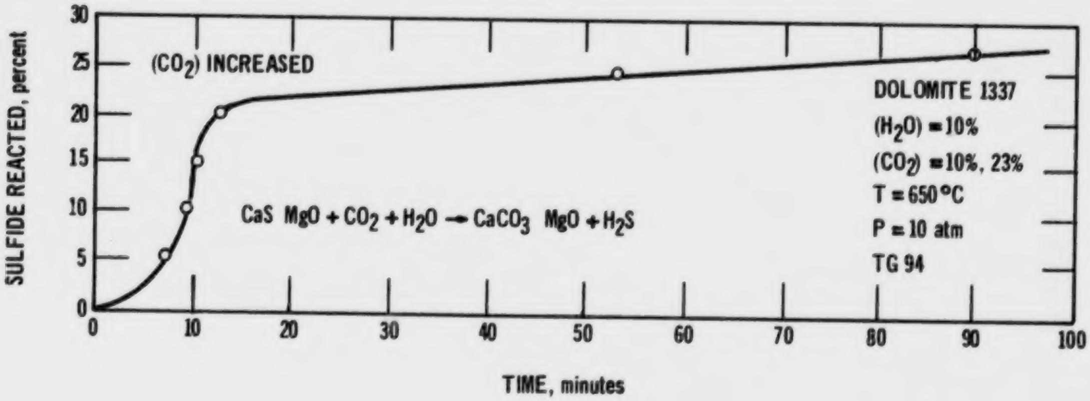

Figure 8. Regeneration of dolomite.

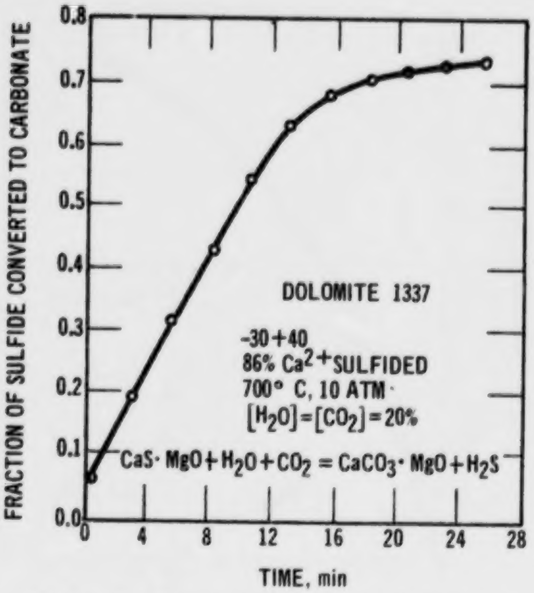

Figure 9. Regeneration of carbonate from sulfided dolomite.

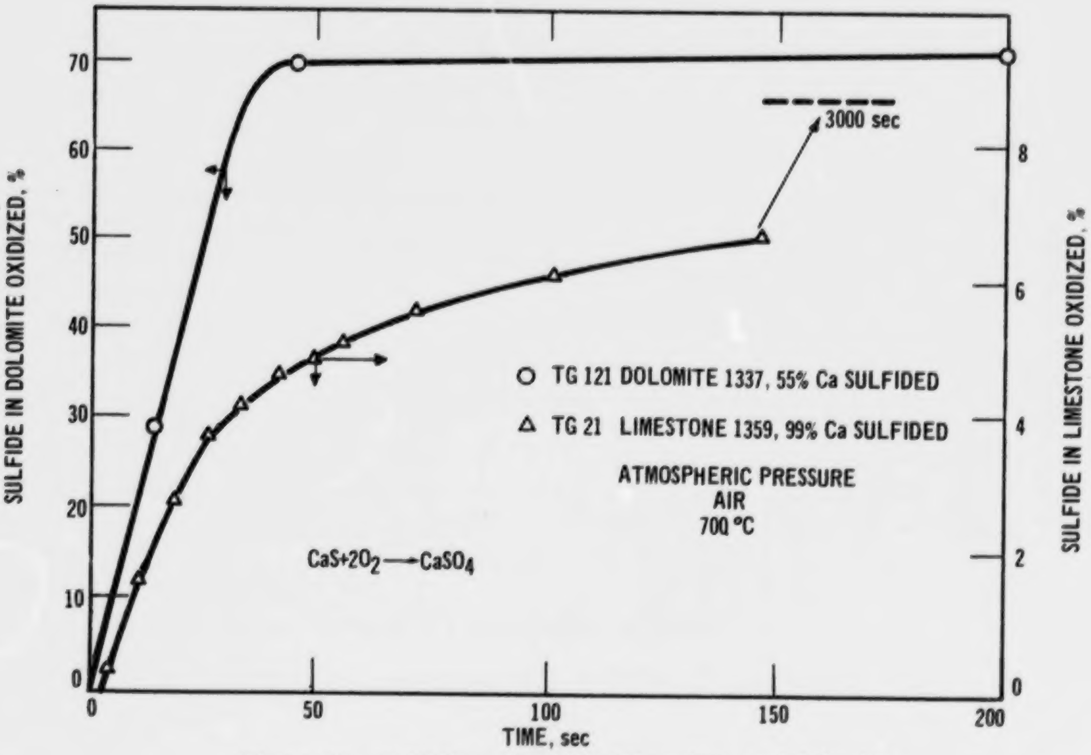

Figure 10. Oxidation of sulfided limestone and dolomite.

SESSION II:

Non-Coal Fluidized-Bed Combustion Processes

SESSION CHAIRMAN

Mr. A. Skopp, Esso Research and Engineering, Linden, New Jersey

1. COMBUSTION IN A CIRCULATING FLUID BED

H. W. SCHMIDT

Lurgi Chemie und Huettentechnik GMBH

INTRODUCTION

To reduce the high heat consumption as compared with the rotary kilns normally still used for calcination of alumina, Vereinigte Aluminium Verke AG (VAW) has developed a fluid-bed process with direct combustion in cooperation with the Lurgi Companies.

The main characteristic of this process, which is generally suited to the application of endothermic processes at low particle diameter, is the circulating fluid bed. Compared with classical fluid beds with constant bed height and a defined surface, a gas/solid mixture of varying concentration in a circulating fluid beds fills up the complete reactor room. The solid discharged at the furnace top together with the combustion gas is fed back to the reactor after being collected from the gas flow in a recycling cyclone. Only by permanent recycling is the system maintained in a constant condition.

This type of reactor has the advantage of passing large gas volumes through relatively small reactor sections.

Since the combustion gas and the fluidization gas are identical, the problem is to optimally coordinate the fluid dynamic conditions for the fluid bed, such as solid recycling and distribution of concentration, with the pyrotechnical requirements, such as mixing and combustion.

CIRCULATING FLUID-BED PILOT PLANT

To investigate these processes, a pilot plant (24 ton/day Al_2O_3) for fundamental studies was erected prior to the construction of the first industrial plant (500 ton/day Al_2O_3). Figure 1 shows a process flow sheet with the most important process steps.[1]

The essential part of the process consists of the calcination circuit which works on the principle of a circulating fluid bed. Optimum thermal efficiency of the whole process is reached by preheating the solid with the combustion gas flow, and the combustion air with the solid discharge flow.

The feed hydrate is preheated by the waste gas flow and partially dehydrated in two Venturi dryer stages. The alumina, which is preheated to about 400°C and has a loss on ignition of 7-8 percent, is subjected to final calcination in the fluid-bed furnace.

The energy required for the endothermic process, water evaporation, dehydration heat, radiation and discharge losses, is supplied by direct combustion of heavy fuel oil in the fluid-bed furnace.

The combustion air is divided into primary air and secondary air. The two air streams are preheated in a multi-stage fluid-bed cooler by the discharged alumina. The secondary air flow, which fluidizes the alumina in the cooler chambers, is preheated directly. The primary air flow, which is passed through tube bundles immersed in the fluid-bed chambers, is preheated indirectly.

Figure 2 shows a sketch of the material balance of a circulating fluid bed. The alumina $\dot{m}_A$ discharged at the furnace top together with the combustion gas is composed of the circulating flow $\dot{m}_R$ and the throughput flow $\dot{m}_D$. The ratio of circulating flow to throughput flow $\dot{m}_R : \dot{m}_D$ is decisive for the mean retention time of the product. The mean retention times of the alumina may be adjusted between about 10 and 60 minutes depending on the intensity of circulations of the circulating solid.

This adjustment of the alumina retention time needs a defined distribution of solid concentration in the axial direction of the fluid-bed furnace, corresponding to the division of primary and secondary air streams. Because of the high gas velocities compared with conventional fluid beds, the problem is to burn the fuel completely by the time it reaches the furnace top. This must be achieved by optimum mixture of fuel and air. The studies mentioned in this paper therefore concentrate on the combustion efficiency at varying conditions for the mixing ratios of fuel and air.

The phase condition of the circulating fluid bed is illustrated in the fluid-bed phase diagram (Figure 3) as a function of the Fr_k number and the Re_k number. The perimeter is determined by parameters K and M. K and M are nondimensional parameters which result from a combination of the Fr_k number and the Re_k number which derive from the fluid-bed phase diagram.

The range shown in the diagram covers the adjusted test conditions by means of the K and M lines. This range is located in the zone of the aggregative fluidization, a transition phase between classical fluid bed characterized by the particulate fluidization of the individual particle, and of pneumatic transport which starts with the limiting characteristic $3/4 \cdot Fr_k^2 \gamma_G/(\gamma_K - \gamma_G) = 1$. The cross-hatched zone between lines $K = 3.52$ and $K = 2.1$ characterizes at a mean particle diameter $d_{km} = 45\ \mu m$ of the alumina used, the phase condition of the lower furnace section whose concentration is determined by the primary air flow. Lines $K = 626$ and $K = 0.264$ result from the minimum and maximum particle diameters, $d_{kmin} = 8\ \mu m$ and $d_{kmax} = 120\ \mu m$. Lines $M = 0.035$ and $M = 1.33$ are determined by the lowest and highest fluidization velocities.

It can be inferred from this illustration in the fluid-bed phase diagram that the fine particles are preferably discharged from the lower furnace section and subjected to a higher amount of circulation than the larger particles, since the K lines of the smaller particle diameters remain above the boundary line for pneumatic transportation. However, it can be demonstrated by tests that no separation of the fine particles occurs during circulation.[3] This means that under appropriate flow conditions all particles come into the range of pneumatic transport. By impact and frictional forces of the particles and by separation of particle clusters, conditions of fluidization may be changed. Thus, the transport condition is delayed at various times and in various places, resulting in inconstant distribution of solid concentration in the axial direction.

MEASURING EQUIPMENT AND TEST PERFORMANCE

The following parameters were varied for studying the combustion process.

1. Primary air velocity, w_p
   This is the velocity referring to the free cross-section in the lower range of the fluid-bed furnace below the secondary air inlet.

2. Impulse of the secondary air flow, I_s
   This is the force by which the secondary air jets penetrate horizontally into the fluid bed produced by the primary air.

3. Mean furnace temperature, T_m

4. Air ratio of the combustion

For the measurements, exhaust openings were arranged in six measuring sections at various levels of the fluid-bed furnace. It was possible to take gaseous and solid samples by two measuring directions arranged perpendicular to each other, with water-cooled bleed-off lances. Figure 4 shows the measuring arrangement for the tests. The alumina sucked off together with the gas is separated in a cyclone. After cleaning and drying, the gas is passed to continually operated gas analyzers for determining the concentration of the components CO, CO_2, and O_2.

The central part of the lance is composed of a thermocouple for ascertaining the temperature prevailing at the corresponding measuring point. By control of the suction pump the exhausted gas rate can be adjusted in accordance with the temperature at the measuring point in the lance section so that isokinetic suction conditions exist.

With this measuring equipment, the following measuring variables can be determined in a radial direction of each measuring section:

1. Gas concentrations of CO, CO_2, and O_2.
2. Temperature, T_m.
3. Concentration of solid, C_M.

INFLUENCE OF SECONDARY IMPULSE ON THE EFFICIENCY OF COMBUSTION

From the radial profiles measured, mean values of the measuring section areas are formed by integration over the furnace cross-section. From the integral mean values of the various section areas, the axial distributions of gas concentrations, solid concentrations, and temperatures can be determined over the height of the fluid-bed furnace.

In Figure 5 the axial temperature distributions are stated for various mean furnace temperatures (T_m). For the greater part of the fluid-bed furnace, the lines show a constancy of temperature which does not occur in conventional fluid beds and can only be explained by the intensive solid circulation. The decrease in temperature in the lower part of the furnace is caused by the solid feed and the remaining dehydration heat of the calcination process still to be applied in the range. Tests at a higher ratio of circulating solid to throughput solid $\dot{m}_R:\dot{m}_D$ show that the temperature drop in the lower furnace section can hardly be observed any longer.

Distributions of solid concentration in axial direction at varying velocities referred to in the free cross-section are represented in Figure 6. In the lower furnace section, the curves show increasing solid concentration C_M (x) at decreasing primary air velocities (w_p). Starting with coordinate X/D = 1.4, the solid concentrations of all curves show a significant decrease with volume flow increase by the secondary air flow ($\dot{V}_S$). Downstream of coordinate X/D = 3.9 the axial distributions of the solid are almost constant up to the furnace top.

To analyze the efficiency of combustion, it is necessary to determine the combustion rate of the fuel components.[4] Except for the local gas analysis values CO, CO_2, and O_2, it was possible to rely upon the unburnt carbon percentage which was analyzed from the sucked off alumina. These deposits of unburnt carbon on the alumina are mainly found in the lower measuring section areas, decreasing in accordance with the course of combustion up to the furnace top.[3]

The active γ-alumina coming from the preheating section at a temperature of about 400°C is also fed to the zone of the fuel which is directly injected into the fluid bed. Due to the catalytic activity of this γ-alumina, the cracking process initiating the combustion of the heavy fuel oil is greatly enhanced. The catalytic effect on the cracking process must be exclusively caused by the activity of the γ-alumina since the remaining alumina, which has a temperature of 1100°C, is no longer active as a catalyst.

On the basis of the burnt carbon percentage, a molar balance of the oxygen required for combustion, and the measured gas analysis values for CO, CO_2, and O_2, it is possible to establish the necessary equations to determine the development of combustion.

From the radially measured values, such as shown for characteristic tests in Figure 7, the distribution of combustion rates in axial direction of the fluid-bed furnace are obtained by radial and axial integration. Figures 8 and 9 show the combustion curves from which the main factors of influence on the degree of combustion can be seen.

Figure 8 shows efficiencies of combustion with constant impulse of the secondary air jets and varying primary air velocities (w_P), which are proportional to the velocity referring to the free cross-section (w_G). At decreasing values of the primary air velocity (w_P), a steeper course of the combustion curves can be observed. This result is attributed to the retention time of the combustion gases in the furnace, which increases at decreasing velocity.

Figure 9 shows percentage of combustion at a constant primary air velocity (w_P), but varying impulses (I_S) of the secondary air jet penetrating into the fluid bed. Although the high primary air velocity (w_P) shortens the retention time of the combustion gases, substantially steeper combustion degrees can be reached at increasing secondary air impulse (I_S) than in the case of curves as per Figure 8. This result is due to the more intensive radial mixing of the fuel components with the combustion air. The radial distributions in Figure 7 in the two different measuring section areas clearly show this influence. At a lower secondary air impulse an increase of the CO_2 concentration exists only in the zone near the furnace wall. At a higher impulse value a homogeneous distribution of concentration over all the measuring section areas exists.[5]

The results show that by suitable distribution of primary and secondary air streams, the combustion reaction can be substantially speeded up. This will permit an increase of the gas throughput and a higher specific load of the fluid-bed furnaces, along with a reduction of the height of the fluid-bed furnace.[3]

In conclusion, it should be mentioned that apart from the combustion process as studied in a pilot plant, two industrial plants, each having a capacity of 500 metric ton/day Al_2O_3, are successfully operating according to the described process; another three plants, each with a capacity of 650 metric ton per day Al_2O_3, are under construction and will be started up in the course of the next year.

SUMMARY

In a pilot plant for the calcination of alumina according to the circulating fluid-bed method, the combustion process is studied experimentally. Since high gas velocities occur in the circulating fluid bed, the retention times of the combustion gases are short at direct combustion in the fluid bed. By a suitable division of the primary and secondary combustion air flows, it is possible to increase the radial mixing and to substantially speed up the combustion of the fuel components. It is thus possible to increase the specific throughput capacity by raising the gas rate.

During the tests, deposits of unburnt carbon on the alumina were observed in the lower furnace section. Consequently, the endothermic process can only be performed according to the principle of circulating fluid bed since the carbon is not completely burned until the fuel reaches the furnace top.

NOMENCLATURE

C_M = Solid concentration, kg/Nm^3
D = Diameter of reactor, m
d_k = Particle diameter, m
Fr_k = Froude number (-)
g = Acceleration of gravity, m/sec^2
g_v = Burned-out portion of fuel (-)
g_u = Unburned portion of fuel (-)
I_S = Impulse of secondary air flow, m kg/sec^2

$$K = \frac{\nu^2}{d_k^3} \cdot g\left(\frac{\cdot \quad \gamma_G}{\gamma_K - \gamma_G}\right)$$

$$M = \frac{w_G^3 \cdot \gamma_G}{\nu \cdot g(\gamma_K - \gamma_G)}$$

$\dot{m}_A$ = Solid output on furnace top, kg/hr
$\dot{m}_P$ = Solid throughput, kg/hr
$\dot{m}_R$ = Circulating solid, kg/hr
Re_k = Reynolds number (-)
T_m = Mean temperature, °C
$\dot{V}_P$ = Primary air flow, Nm^3/hr
$\dot{V}_S$ = Secondary air flow, Nm^3/hr
$\dot{V}_F$ = Waste gas flow, Nm^3/hr
w_G = Superficial gas velocity, m/sec
w_P = Superficial velocity of primary air, m/sec
L_M = Efficiency of combustion, gv/gu + gv)
γ_G = Specific gravity of gas, kg/m^3
γ_K = Specific gravity of solid, kg/m^3
ν = Kinematic viscosity of gas, m^2/sec

BIBLIOGRAPHY

1. Reh, L. Fluid Bed Processing. Chem. Eng. Progr. 67(2). 1971.
2. Ernst, J., L. Reh, K.H. Rosenthal, and H.W. Schmidt. Experience with the Calcination of Aluminum Trihydrate in a Circulating Fluid Bed. (Presented at American Institute of Mechanical Engineers Meeting. New York. Paper number A-71-4. February 1971.)
3. Schmidt, H.W. Uber den Verbrennungsverlauf in zirkulierender Wirbelschicht. Dissertation, Karlsruhe 1971.
4. Gunther, R. Ausbrand von Strahlflammen. Archiv f.d. Eisenhüttenwesen. 39(7):515-519, 1968.
5. Martsevoi, E.P. Spread of a Gas Jet in a Cross-Flowing Stream of Different Density. Gas-Institute, Acad. of Sciences USSR, translated from Teoreticheskie Osnovy Khimicheskoi Technologie. 3(4):644-646, 1969.
6. Reh, L. Das Wirbeln von körnigem Gut im schlanken Diffusor als Grenzzustand zwischen Wirbelschicht und pneumatischer Förderung. Dissertation, Karlsruhe 1961.

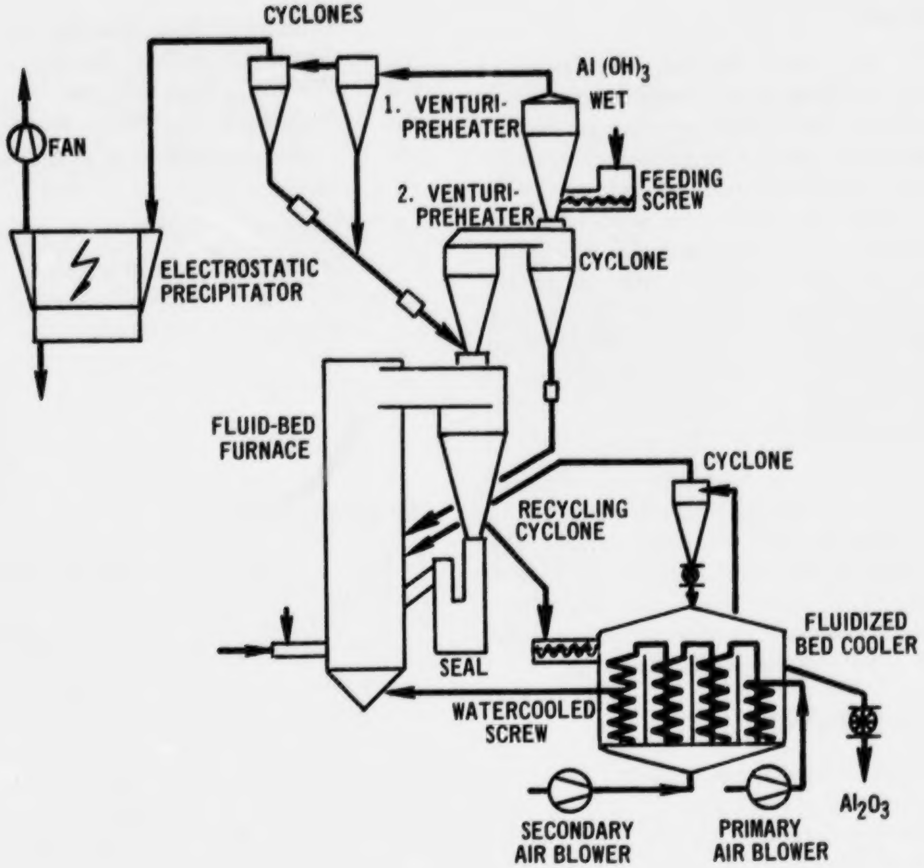

Figure 1. Flowsheet of fluid-bed calcining plant.

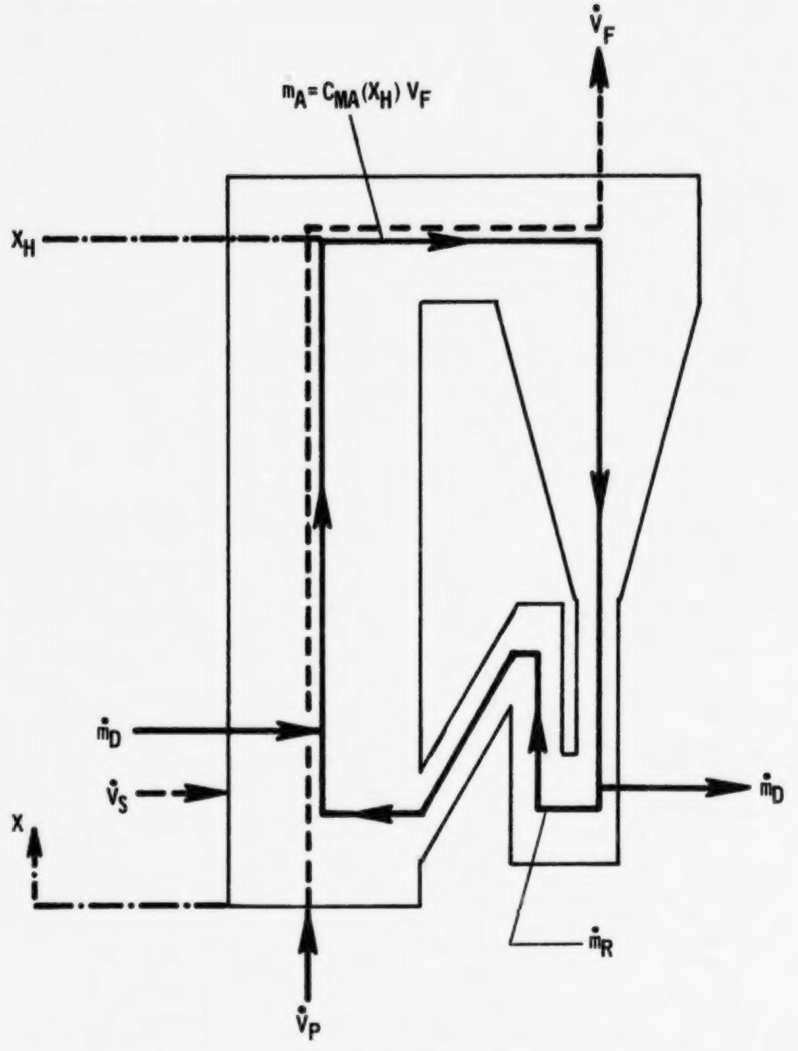

Figure 2. Material balance of circulating fluidized bed.

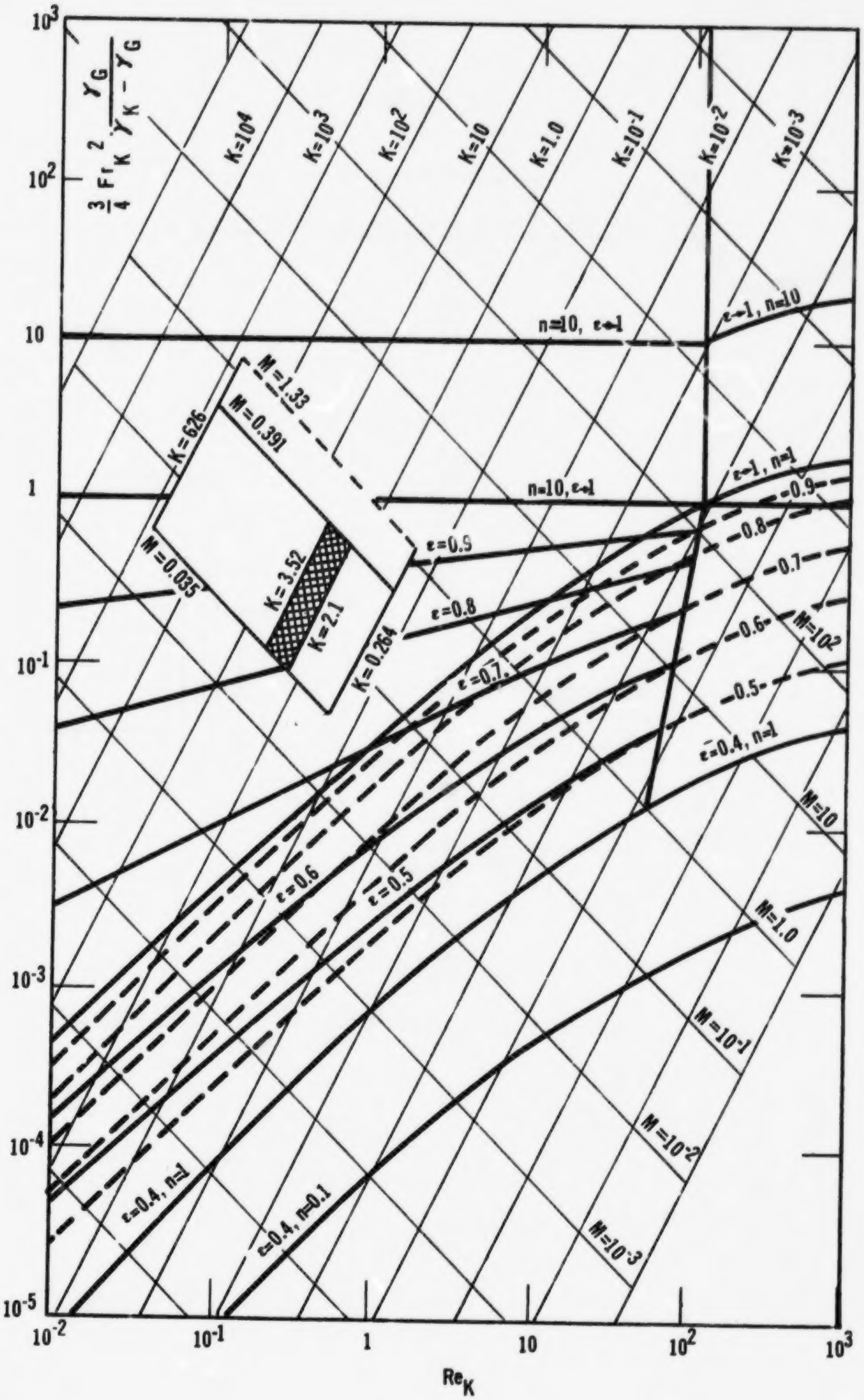

Figure 3. Fluid-bed diagram.

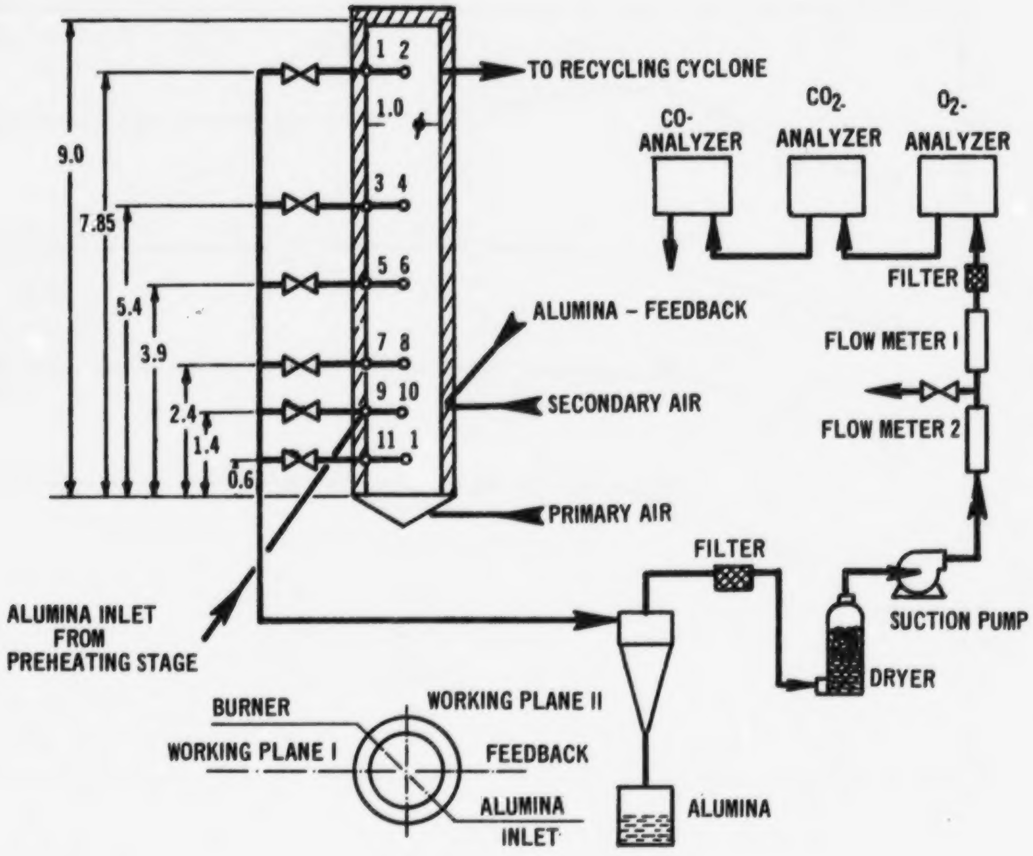

Figure 4. Measuring arrangement.

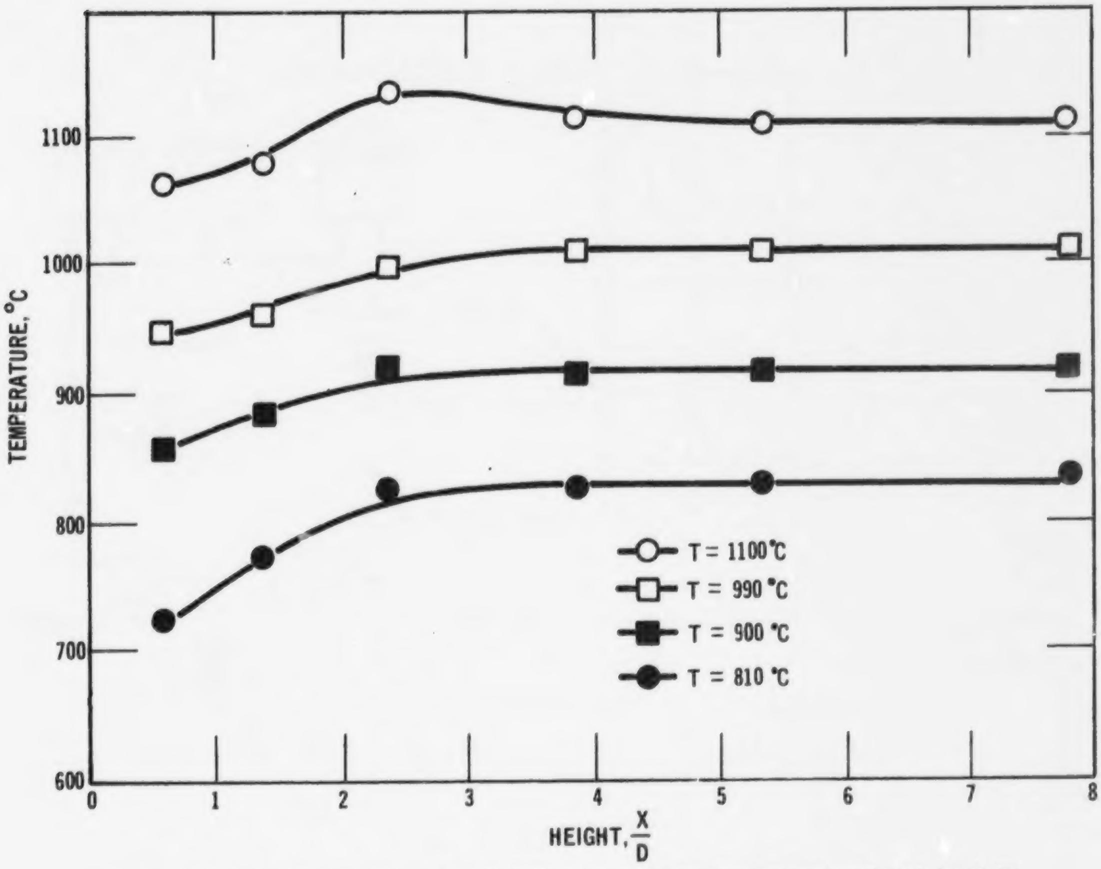

Figure 5. Axial temperature T_m distribution versus nondimensional height X/D of fluid-bed furnace.

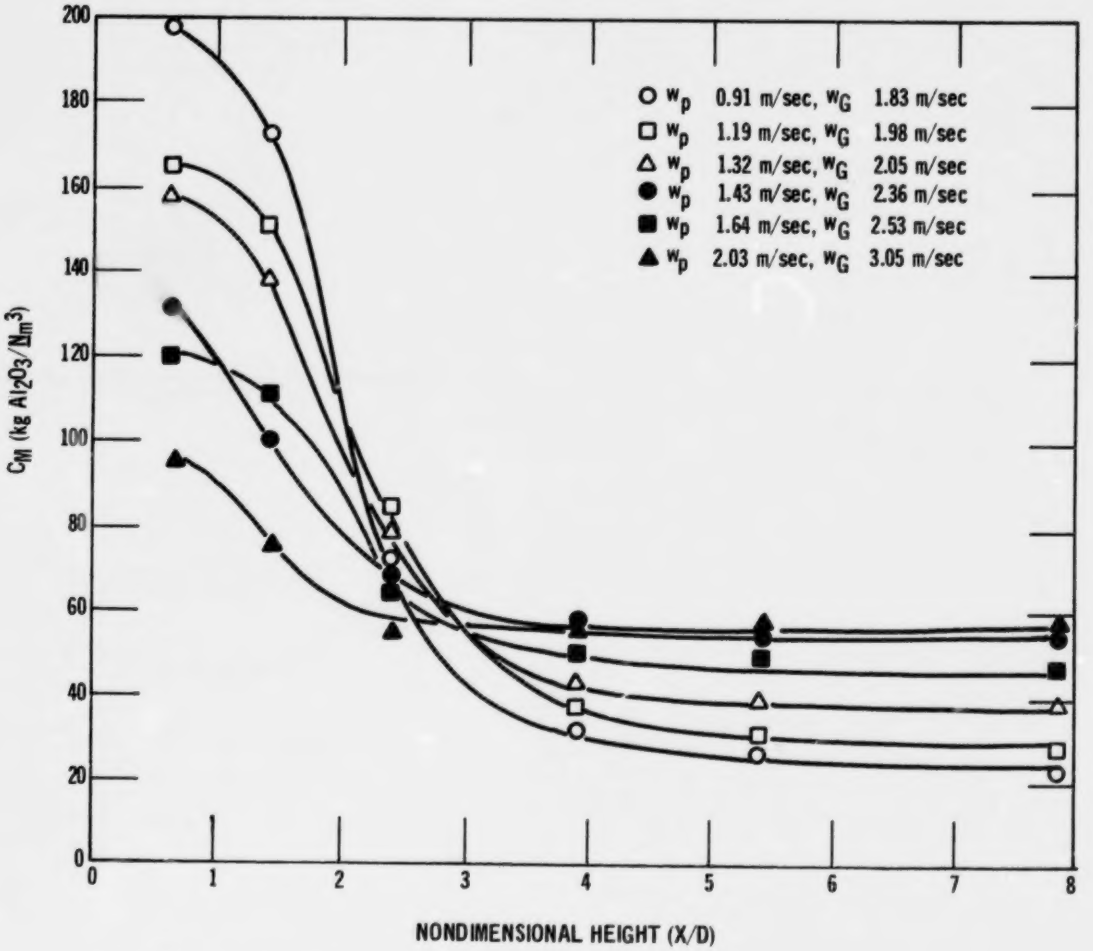

Figure 6. Axial solid concentration (C_M) distribution versus nondimensional height ($X/_D$) of fluid bed furnace.

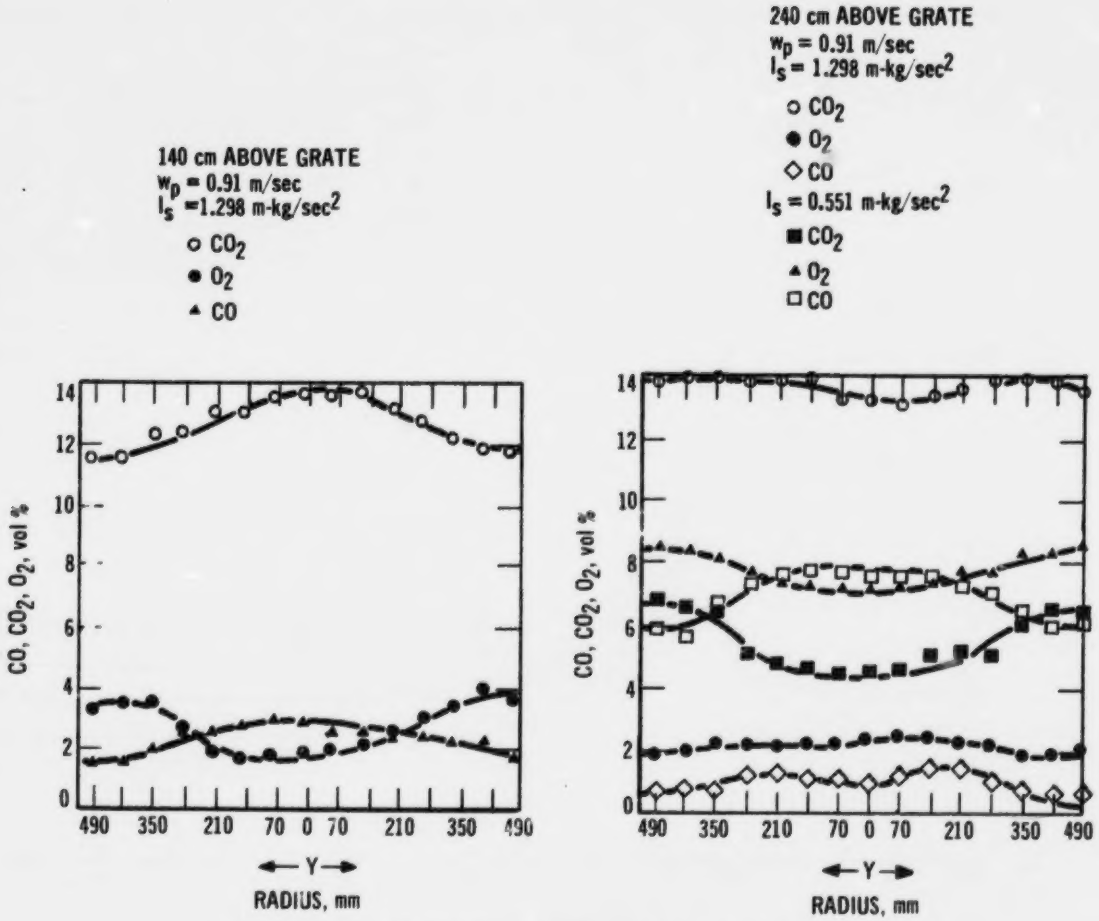

Figure 7. Radial distribution of gas concentration 140 cm and 240 cm above grate at constant primary air velocity (w_p) and variable secondary impulse (I_s).

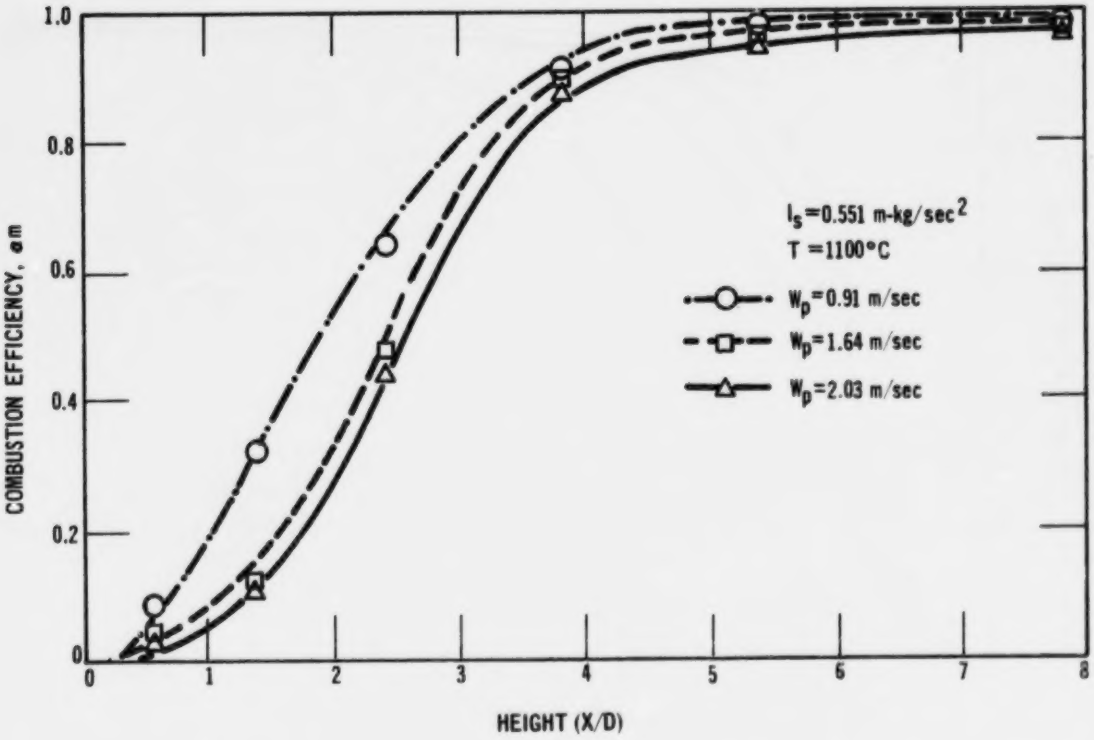

Figure 6. Efficiency of combustion (α/m) at constant impulse (I/s) and varying primary air velocities (w/p).

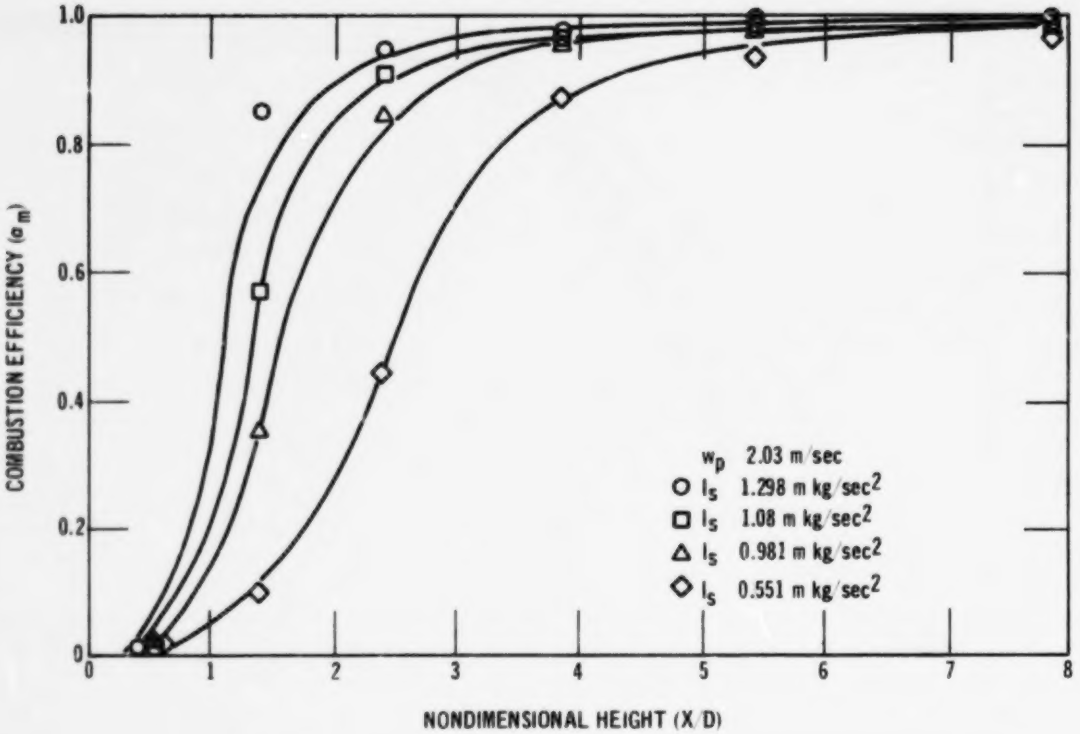

Figure 9. Efficiency of combustion (αm) at constant primary velocity (W_p) and variable secondary air impulse (I_S).

2. DISPOSAL OF SOLID WASTES BY FLUIDIZED-BED COMBUSTION

G. G. COPELAND

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ABSTRACT

Those of us who labor in the field of pollution control have grown to realize that solid waste disposal is the largest pollution problem which society must face in the coming years. For too long, we have left the problem to municipal fathers, who for one reason or another have either ignored the problem or have installed equipment which has long since been made obsolete by modern technology.

We hear every day about the virtues of land filling garbage and using municipal sewage sludge as soil conditioner, "a la night soil" techniques used in the Far East, because incineration is too expensive, too dirty, makes smoke, and needs high chimneys. This thinking is obsolete in the face of present day fluid-bed technology which is substantially less costly than conventional incinerators, is not dirty, and cannot be made to smoke if operated properly. To our knowledge, there is not a fluid-bed system in operation anywhere in the world which is backed up with a smoke stack.

This paper covers the development of the largest fluid-bed solid waste incinerator in the world — an installation which we feel is the forerunner of the next generation of solid waste disposal systems and the best means of solving the solid waste disposal problem.

INTRODUCTION

Having been one of the midwives at the birth of fluidized-bed technology in the 1940's, it is a pleasure for me to be here today at this Third International Conference on Fluidized-Bed Combustion, and to be able to discuss the technology with so many people who are knowledgeable in its applications.

As a matter of fact, I was one of the first persons assigned the responsibility of selling fluid-bed technology outside of the oil refining industry.

Looking back on those years of trying to convince the technical world that we weren't crazy, and at the same time having to show sales progress while doing research all day, all night and weekends, it is a wonder that those of us who were engaged in this exciting development did not throw up our hands in disgust and go to something more certain.

In those days when the new applications were basically metallurgical, it seemed that all fluid-bed installations would always be in out-of-the-way places like Red Lake, Ontario; Arvida, Quebec; or Berlin, New Hampshire. For sure, however, all startups over a five-year period were in the dead of winter, and could only be described as miserable.

Today, our startups are in places like Florida, Japan, South Africa, the Bahamas — as always, in the winter time; but the climate is certainly much better on the average, and we have the technical understanding and support of people who have learned something about the technique.

The technology has come a long way in the intervening 25 years, and I believe it is now coming into its own with new and exciting applications popping up all over the world. Having been active in the business throughout these years, I think my greatest satisfaction comes from meeting young engineers, fresh out of school, who have been taught at least the basic elements of the techniques and can go on to understand the many permutations and possibilities of fluid-bed processing.

The company, which bears my name, has specialized in air and water pollution control since the early 1960's and uses fluid-bed combustion technology as the heart of many patented processes which the company sells on a worldwide basis.

We now have fluid-bed waste disposal combustion installations in virtually every major industry, many on a first time basis, and have successfully used the technique on both liquid and solid waste materials.

We like to think that in developing certain fluid-bed processes to dispose of pulp and paper mill wastes, we did accomplish the impossible by burning organic matter in a fluid bed in the presence of inorganic salts having a low fusion point. We did, in fact, what most of the early publications on fluid beds said was impossible.

Our first such commercial installation was in pulp and paper pollution control, where we were required to destroy the organic matter removed from wood pulp in an inorganic solution of sodium-sulfur salts. We knew that a fluid-bed temperature in excess of 1300°F was necessary to get complete burn-out of organic (polluting) matter; at the same time we knew that the inorganic salts would fuse at 1350 to 1370°F depending on the inorganic mix.

By taking advantage of the partial fusion of inorganics, we were able to force pelletization of the inorganics to form the fluid-bed medium; our systems actually operate easily between 1300 and 1350°F. We have several such plants which have been in successful operation for over 10 years — providing a solution to pollution problems which were considered impossible in the 1950's.

For the most part, our early development of fluid-bed technology for pollution control dealt with waste solutions. In 1971, however, we built what we believe is the largest solid waste fluid-bed incinerator in the world at Great Lakes Paper Company in Thunder Bay, Ontario. We believe this installation broke some new ground in solid waste disposal by fluid-bed technology. This paper covers that installation in some detail.

FLUID-BED INCINERATORS IN GENERAL

The principle of fluidizing solid materials at elevated temperatures in the presence of and by means of a gas, was first commercially developed by the oil refining industry in the form of fluid-bed catalytic crackers. While fluid-bed incinerators only vaguely resemble "cat crackers" they do function because solid particles are set in fluid motion (fluidized) in an enclosed space (fluid-bed zone) by passing combustion air through the fluid-bed zone in such a way as to set all particles in that zone in a homogeneous boiling motion.

In this state, the particles are separated from each other by an envelope of the fluidizing gas (air for combustion) and present an extended surface for a gas to solid reaction, as for example, air to carbon-hydrogen. This extended combustion surface makes possible the high thermal efficiency found in most fluid-bed reactors.

Capacity is a function of reactor bed total area, but is usually expressed as fluid-bed surface area.

At combustion equilibrium, the fluid bed resembles a boiling liquid and, in fact, obeys most of the hydraulic laws. The dispersion of fluidizing gas throughout the fluid-bed zone by the specially designed orifice plate, assures complete mixing; temperature variations from any one spot in the fluid bed to any other will not normally exceed 10 to 15°F.

The mass of fluid-bed medium is kept at combustion temperature by oxidation of the organic material in the feed by the oxygen contained in the fluidizing air. There is little or no flame, but rather a glowing condition. Combustion is virtually instantaneous and the fluid-bed proper will contain no unburnt organic material. Complete oxidation is the key to the control of air pollution.

It is fundamental to any incineration process that there be no air pollution problem resulting from incomplete combustion of waste solids. In this respect, fluid-bed units are superior to any other type of combustion. This is again due to the extended surface presented by the fluid-bed medium. Copeland designed units normally will burn in excess of 300,000 Btu/ft^2-hr. This could be compared, for example, with coal burning boilers which do well to consume 40,000 Btu/ft^2 of grate area/hr.

A commercial unit burning sewage sludge at 1400°F had the stack gas analyses shown in Table 1.

Table 1. MASS SPECTROGRAPHIC ANALYSES[a]

Component	Sample number 1	2	3
Volume, %[b]			
CO_2	5.17	8.33	8.86
A	0.73	0.07	0.60
O_2	14.4	9.2	7.4
N_2	79.7	81.8	83.0
Volume, ppm[c]	ND	ND	ND
SO_2			
COS			
H_2S			
NO_x			
Hydrocarbon			

[a] Dry basis.

[b] Samples were taken at 11:30 a.m., October 30, 1968.

[c] Not detectable by mass spectrographic analysis.

Every installation built in the last ten years is meeting the air pollution regulations of the state in which it is located and complies with the most stringent air pollution regulations of the country. This applies to both gaseous and particulate matter in exhaust gases.

Perhaps the most important and least understood feature of Copeland incinerators is the ability to pelletize most inorganic ash residues to form the fluid-bed medium itself. Many fluid-bed incinerators use sized sand as the fluid-bed medium, but the presence of low melting point inorganics makes pelletization a useful technique which in some cases permits the recovery of a saleable product uncontaminated by sand or other diluents. Pelletized ash is dust free and easy to handle. For this reason, where possible, we endeavor to force pellet growth in our incinerator systems.

In every case of forced pelletization, we find reduced dust collection problems and a generally easier operation. Pellet growth is a function of temperature and surface area in the bed itself; its rate is controlled by controlling both temperature and unit area in the bed. By occasional screen analysis of the bed product, we can predict rate of growth and adjust it to the needs of the system.

Fluid-bed systems capacities are a function of superficial space velocity or the rate at which the fluidizing gas is forced through the fluid bed. A unit designed for a 2 ft/sec space velocity will give 100 percent additional capacity if enough air is forced through it to raise the velocity to 4 ft/sec, provided the velocity is not sufficient to elutriate the bed material out of the reactor. Space velocities are chosen to meet a given set of feed conditions and will generally be in the range of 1.0 to 5.0 ft/sec. Installations have been built for other purposes where the velocity has exceeded 10 ft/sec.

Of great importance in basic design is the opportunity to build into fluid-bed systems a future capacity at minimal cost. By installing a false brick lining in the fluid-bed zone at time of original construction, combustion area is provided for future use. For example, a project requiring a 10-ft diameter reactor can get 30 percent future capacity built in by adding a 1-ft thick false lining, for less than a 5 percent increase in present cost.

DESIGN CONSIDERATIONS FOR FLUID-BED INCINERATION PROCESSES

Fundamental to the design of any fluid-bed unit is a clear understanding of the waste material to be processed. Fluid beds will not work on material too coarse in size to be fluidized or having an ash content with a lower melting point than the temperature necessary for complete oxidation of the organic matter. In this latter case, fluid beds do not differ from the older type incinerators.

Generally it can be said that fluid beds will burn anything that can be fed into them and fluidized. Solid wastes with a minimum of free surface water are generally blown into the reactor, whereas drier materials can be fed by means of a sealing type screw conveyor. Semi-plastic sludges such as sewage sludge are fed by screw conveyor or more simply by a progressing cavity pump. Thermoplastic materials like grease are most readily fed by first being melted and then pumped by centrifugal pump.

Wherever possible in our design, we try to build into the system enough freeboard residence time to permit some heat exchange between the incoming wet feed and the outgoing combustion gases. Since these gases are generally wasted, evaporation of water from the incoming feed by direct heat exchange has the effect of improving thermal efficiency.

Before designing any fluid-bed system, we pay particular attention to the chemical composition of the material and look for trace elements which might have a fluxing or fusion point lowering effect on the ash content. We have found, for example, that a fluid bed burning organic material, the ash content of which is 100 percent sodium chloride, can be operated in excess of 1300°F without fusion problems. Yet another product of the same type having 1.5 percent sodium chloride could not be burned in excess of 1150°F without complete fusion. Obviously, this is a typical eutectic problem, but critical in any incinerator design.

VIRTUES OF FLUID BEDS USED IN SOLID WASTE INCINERATION

The higher combustion efficiency of fluidized beds is attributable to a number of melting point inorganics makes pelletization a useful technique which in some cases permits the recovery of a saleable product uncontaminated by sand or other diluents. Pelletized ash is dust free and easy to handle. For this reason, where possible, we endeavor to force pellet growth in our incinerator systems.

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VIRTUES OF FLUID BEDS USED IN SOLID WASTE INCINERATION

The higher combustion efficiency of fluidized beds is attributable to a number of characteristics found only in part in other combustion techniques. These may be described as follows:

Extended Surface

Total surface area, in or on which the combustion process takes place, is a very important design consideration.

We have a commercial installation burning 60 x 10^6 Btu/hr (waste sulfite liquor) which has surface area in the bed medium (90 tons) equivalent to the surface of a super highway 70 miles long. Bed medium in this case is pelletized inorganic salt recovered as a by-product of combustion. Pellet size is basically 14 to 65 mesh Tyler screen scale.

Obviously, the more surface area, the better the opportunity for reaction between oxygen in the fluidizing air and combustible material.

Residence Time

Combustion is a time-temperature reaction which is most efficiently carried out under conditions which give instantaneous reaction. Lack of time or temperature will make for incomplete reaction and produce partial products of combustion.

In systems where temperature must be controlled at lower limits because of other thermal considerations, residence time therefore becomes an important factor. We have noted in systems burning waste sulfite liquor at 35 percent solids that combustion at 1300°F is instantaneous with no residual carbon left in the bed. However, at 1250°F combustion is slow and, if allowed to proceed for any length of time, carbon build-up in the bed is noticeable.

TYPICAL APPLICATIONS OF FLUID-BED INCINERATORS

Fluid-bed incinerators are finding application in the combustion of waste solids and liquids because in many cases these waste materials could not economically or practically be incinerated by older more conventional equipment. The very fact that many waste solid materials have been used for land fill, rather than completely destroyed by combustion, is usually an indication of some difficulty with conventional processes.

Since disposal usually implies an outright cost to the producer, the most efficient system must be found; fluid beds are being chosen because of higher thermal efficiencies, better control of odors and particulate matter emissions, and a simpler process having greater design latitude.

Fluid-bed incinerators have been demonstrated by commercial practice to be readily applicable to the combustion of the following types of solid waste materials:

1. Domestic sewage sludge.
2. Municipal garbage.
3. Oil refinery wastes such as:
    API separator sludge,
    tank bottoms,
    waste caustic streams, and
    general refuse.
4. Petrochemical wastes such as:
    hydrocarbon compound sludges, and
    complexed waste inorganics.
5. Water treatment plant carbonate sludges.
6. Packing house wastes.
7. Distillery slops.
8. Pharmaceutical plant wastes.
9. Clarifier effluents from most industries.
10. The destruction of lethally poisonous materials.
11. Pulp and paper mill sludges and various solid wastes.

The foregoing list is by no means complete; it will be seen that some waste materials could be destroyed by older techniques. However, the high thermal efficiency of fluid beds makes it possible to incinerate these materials at much higher water contents without the use of extraneous fuel, thus giving fluid bed incinerators the nickname "water burners."

FLUIDIZED BEDS VERSUS POLLUTION CONTROL

Water Pollution

Combustion of waste material by fluid-bed techniques is the ultimate of disposal techniques in that no liquid effluent need result. Ash produced by fluid-bed combustion is completely burned out, impresses no BOD if used for land fill, and many inorganic ashes can be reused chemically. Scrubbing of exhaust gases for air pollution control is a necessity; but in most cases we scrub with the waste liquid being combusted and hence produce no new effluent.

Air Pollution

Most fluid beds used in industrial waste disposal use forced air fluidization which permits an easier route to high pressure drop treatment of exhaust gas. We usually install dry cyclones after the reactor and take up to 15 inches of water drop across them. These are generally followed by wet scrubbing where up to 50 inches of water pressure drop are taken.

We find that these systems, although costly in terms of power consumption, are exceeding by as much as 50 percent the most stringent air pollution regulations now in effect. Our experience indicates that, if sub-micron particulate matter is to be taken out of exhaust gas, high pressure drop across scrubbing equipment is necessary. If sub-micron particles escape the scrubber, a tail gas plume will persist in the atmosphere. Any persistent plume visible to the public is an open invitation to investigation by pollution control authorities.

Regardless of present air pollution regulations, our experience tells us that the ultimate regulation will demand no visible plume whatsoever; this may include water vapor plumes as well.

We believe that fluid-bed combustion systems, properly designed and incorporating the newest scrubbing techniques, offer the best answer to ultimate air pollution regulations.

A recent trend in municipal sludge incineration, promoted by new EPA regulations, will require that exit gas from sludge incineration be exposed to a temperature of 1600°F for 2 seconds to destroy malodorous gases and chlorinated hydrocarbon compounds which are produced by incineration at lower temperatures. This will make it impossible to dispose of municipal sludge by older incineration methods without fuel burning after-burners.

WORLD'S LARGEST SYSTEM FOR BURNING BARK, DEBRIS, AND SLUDGE

In December of 1971, we brought on stream a solid waste, fluid-bed incinerator, which disposes of about 600 ton/day of pulp and paper mill wet wood waste at a water content of 65 to 70 percent without the use of extraneous fuel. The composition of the feed is given in Table 2.

The Great Lakes Paper Company Limited at Thunder Bay, Ontario, had two solid waste disposal problems.

The first problem was a huge pile of bark (300,000 yd^3) which had been accumulating for years, and was beginning to make its presence felt by spilling over into the local river. The second problem arose from the installation of clarifiers on the effluents from the groundwood mill, the sulfite mill, and the Kraft mill, the sludge from all of which would have to be disposed of.

A conventional boiler was already in use at the mill for burning bark fresh from the barking drums. But (apart altogether from questions of existing capacity) disposal of the clarifier sludge and pile bark by such conventional means would require that they be further dewatered. The old piled bark, however, contained all manner of junk, including a generous proportion of stones, which represented a potential source of damage to existing bark presses. And the clarifier sludge, on account of its slimy, fibrous nature, was very

Table 2. PROPERTIES AND DAILY QUANTITIES OF FEED TO COPELAND SYSTEM

	Dry solids			
	%	Ash, %	Btu/lb	Ton/day
Sludge feed				
Sludge from groundwood clarifier	25	2.2	7100	40
Sludge from kraft clarifier	25	2.3	7155	5
Rejects from groundwood mill	25	1.9	7155	5
Rejects from kraft mill	25	1.9	7155	5
Average properties and total amount	25	2.2	7155	55
Wood waste feed				
Bark and wood debris	35	2.0	8500	50
Surplus bark	35	2.1	8500	20
Average properties and total amount	35	2.0	8500	70

hard to dewater over 25-28 percent solids. At this low consistency, it would cause combustion troubles in the conventional bark burning furnace.

Consequently, Great Lakes' management turned to the Copeland fluidized-bed process, which offered as one of its characteristics the ability to burn woody materials with as little as 30 percent solids without supplementary fuel. This would make it possible to burn, self-sufficiently, the unpressed bark and/or a mixture of the unpressed bark and clarifier sludge. A Copeland unit of 180 BD ton/day capacity was therefore decided on.

The next question was whether this unit should be attached to a waste heat boiler for raising steam. It was recalled, however, that 24,000 lb/hr of steam was currently being used to heat 4000 gal/min of hot water for woodroom showers. Since this heat demand was of the same order as that expected to be available by recovery from the Copeland unit, it was decided to produce hot water directly in the unit's heat recovery-scrubbing system. In this way, the mill steam supply available for other uses was increased, without going to the expense of installing another waste heat boiler.

Bark Feed

The bark from the old pile and other wood debris is picked up in the mill yard by truck, and combined with slasher sawdust, groundwood snipes, and wood scraps in a surge hopper. From here the waste is mechanically conveyed to a wood hog, which breaks it into fragments which can be handled efficiently by the subsequent pneumatic conveying system (smaller than 6x6x6 inches). The shredded waste resulting is then transferred by bucket elevator to a storage silo. This silo holds 1 day's feed to the system, so that a man for collecting bark and wood debris is needed on one shift only. The silo has a live-bottom vibrating hopper, which discharges the waste through a vibrating feeder into a pneumatic conveyor which injects it into the reactor immediately above the fluid bed.

Stones up to 6-in. diameter in the bark and wood debris, are conveyed along with wood waste to the fluid-bed reactor by a pneumatic feeder. Tramp metal is removed by an electromagnet.

Sludge Feed

The wet, fibrous clarifier sludge is pneumatically conveyed to the disposal system from the sludge collecting tanks, after dewatering by filtration. Though it could be injected directly into the reactor above the fluid bed, the large amount of air required to convey it would, under these conditions, enter the reactor and critically lower its operating temperature. Normally, therefore, the sludge is separated from its conveying air in a cyclone from which it is mixed with the normal wood wastes.

Combustion System

The fluid-bed reactor, is a carbon steel vessel lined with insulating and refractory brick. A five-stage, centrifugal, low-pressure blower supplies the fluidizing and combustion air. The fluidizing air is distributed into the bed by an orifice plate separating the windbox from the bed section. The fluid-bed zone contains the fluidizing medium, which is made up of sand removed from the waste wood.

Above the bed zone, the reactor widens out to form the disengagement or freeboard zone. The increase in reactor diameter here is sufficient — even in view of the increase in volume of gas phase due to the generation of water vapor — to reduce the upward velocity of the gases so that particulate solids will disengage and fall back into the fluidized bed.

Overbed burners are provided for startups as necessary. Auxiliary fuel is not required when the system is fed waste materials at 75 to 120 percent of design capacity.

Sand Handling System

It is necessary, from time to time, to withdraw excess sand and grit from the bed. This is done by discharging it into a sealing screw conveyor, and then to a storage silo.

Gas Scrubber & Hot Water Generator

The hot combustion gases leaving the reactor at about 1600°F pass into a two-stage scrubbing system.

The first stage is an adjustable wetted-throat venturi scrubber, in which the scrubbing water is introduced over a weir and atomized by the energy of the venturi. The resulting fine droplets contact the ash contained in the combustion gases and are separated from the gas in the separator section. The secondary scrubber consists of two beds of fluidized packing, on which scrubbing water is sprayed, thus trapping any residual particulate matter while at the same time picking up heat. Finally, the gases pass through a demister to remove entrained fine droplets before being vented to atmosphere. Since no particulate matter, objectionable gases, or odor have been detected in these exit gases, a high stack to disperse them has not been found necessary.

The scrubbing water from both first- and second-stage scrubbers is collected in an 8500 gallon reservoir below the separator section, from which it is recycled through a pump to the two stages of scrubber trays. The recycle line is provided with a purge line for maintaining the desired temperature and ash concentration in the scrubber liquid. From this reservoir is taken 4000 gal/min of hot water at 155°F used in the woodroom.

Operation & Control

One man operates the plant from a central control panel. During startup operating temperature is reached with auxiliary fuel; when the feed is ignited the auxiliary fuel is shut off. Shut down is accomplished by shutting off the waste feed and the blower furnishing the fluidizing air. The sand bed normally loses less than 100°F/day, so that the unit can be started up without auxiliary fuel after being down for as long as 6 days.

Cost of the fluid-bed installation was approximately $1 million. Labor required is 1.3 man-days/day, and maintenance costs are expected to be low. The operating credit of 24,000 lb/hr of steam is reckoned at $100,000/yr. A further credit is the valuable land that will become available when the old bark pile has been disposed of. Present feed to the unit is 125 ton/day, but short runs have shown that 180 ton/day is feasible. The Air Management group of the Ontario Department of Energy and Resources has monitored the system thoroughly, and has found the exit gases to contain no particulate matter or objectionable gases.

SUMMARY

In bringing this large fluid-bed solid waste incinerator into operation, we have made some interesting discoveries which lead us to believe that the same technique can be used on such other solid waste disposal problems, such as garbage, etc.

To our great surprise, we have found the reactor able to accept large numbers of stones up to 6-in. in diameter, non-magnetic scrap metal, cans, and a variety of things from shredded truck tires to welding rods.

We are fluidizing bed rock particles up to 1-in. cubes and still maintaining good bed stability.

The unit has never produced the first wisp of smoke; air pollution control is so good that pressure drop across the double scrubbing system has been reduced from 50 in. H_2O to 15 to 20 in. H_2O, while still meeting the very stringent air pollution regulations of the Province of Ontario.

The ability of the unit to dispose of large pieces of solid waste has encouraged Great Lakes Paper to institute a change of feed flow which will eliminate the hog completely. When done, all waste, including waste pulp wood sticks up to 4 ft. long will be fed directly into the bed without any size breakdown.

The installation argues well for uncomminuted garbage incineration by fluid-beds — a development which is sorely needed today.

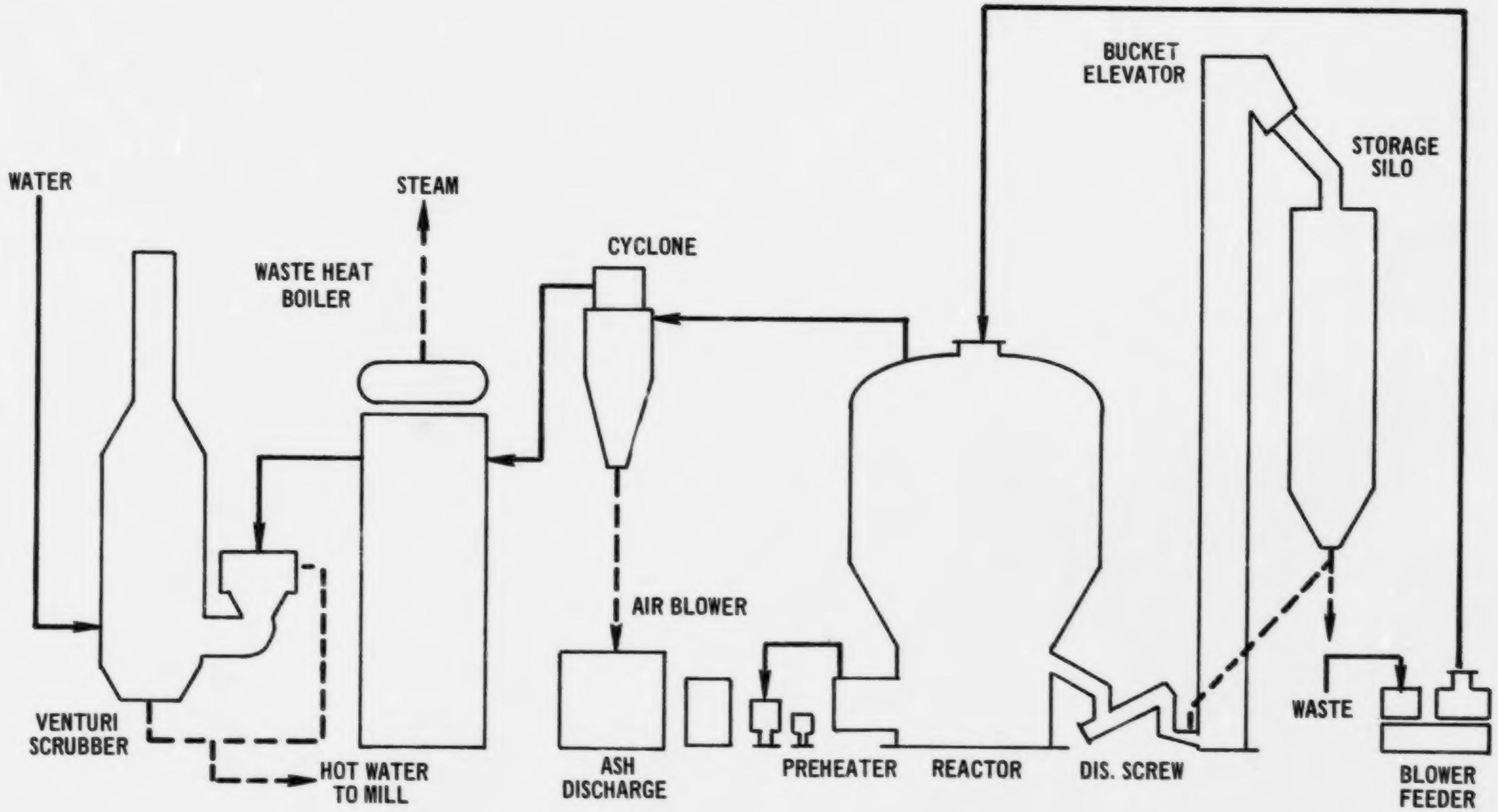

Figure 1. Copeland solid waste incinerator system.

3. FLUIDIZED-BED COMBUSTION OF MUNICIPAL SOLID WASTE IN THE CPU-400 PILOT PLANT

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INTRODUCTION

A series of experiments is currently being carried out to develop a large, high pressure fluidized-bed combustor for incineration of municiple refuse. This publication is based on work performed under Contracts PH 86-68-198 and 68-03-0054 with the National Environmental Research Center, Cincinnati, Ohio, of the Environmental Protection Agency.

The solid waste management concept known as the CPU-400 is sized to accept 400 ton/day of solid waste from municipal packer trucks. Solid waste will be conveyed from receiving pits directly into shredders which discharge into the air classifier units. Components such as metal and glass with high weight to aerodynamic drag ratios will be separated out and conveyed to ancillary disposal or recycling processes. Lighter materials, predominantly papers and plastics, will be transported to a storage conveyor of sufficient capacity to provide a continuous supply of combustor fuel at a uniform rate.

The shredded and classified solid waste is fed into the fluidized-bed combustor through high pressure air-lock feeder valves and a pneumatic transport line. In the fluidized bed, inert sand-sized particles are buoyed and mixed by an upward flow of air coming from the compressor. Heat released by solid waste combustion will maintain the fluidized bed and exiting gas products between 1300 and 1700°F.

Several particulate removal stages will operate on the hot combustion gases prior to their admission into the gas turbine. Inert granular residue will be removed from the fluidized bed and particle collectors as required.

The economic basis for the CPU-400 lies primarily in the recovery of the energy contained in solid waste by virtue of its high content of paper, plastic, and wood products. The recovery and sale of electric power from the disposal of solid waste markedly reduces the cost of the operation. Depending on the value of electrical power and other local conditions, estimated net operating costs range from 2 to $5/ton compared with current incinerator costs of 8 to $14/ton. Where a stable market exists for other reusable materials such as metals or glass, the CPU-400 will also permit recovery of these resources; the process acts to concentrate these recyclable materials by removing the large volume of combustible materials. Revenues derived from recycling will serve to further reduce net operating costs.

The CPU-400 is now in the early stage of its development; system studies and subscale experiments have been completed, and development of the pilot plant is well under way. The fluidized-bed combustor and particulate removal stages currently in development testing are components of the pilot plant.

ADVANTAGES OF FLUIDIZED-BED COMBUSTION

The fluidized-bed reactor is a relatively new approach to the design of high heat release combustors. The primary functions of the air-fluidized inert bed material are to promote dispersion of incoming solid fuel particles, heat them rapidly to ignition temperature, and to promote sufficient residence time for their complete combustion within the reactor. Secondary functions include the uniform heating of excess air and the generation of favorable conditions for residue removal.

The fluidized-bed reactor greatly increases the burning rate of the refuse for three basic reasons:

1. The rate of pyrolysis of the solid waste material is increased by direct contact with the hot inert bed material.
2. The charred surface of the burning solid material is continuously abraded by the bed material, enhancing the rate of new char formation and the rate of char oxidation.
3. Gases in the bed are continuously mixed by the bed material, thus enhancing the flow of gases to and from the burning solid surface and enhancing the completeness and rate of gas phase combustion reaction.

A significant advantage of the fluidized-bed reactor over conventional incinerators is its ability to reduce noxious gas emission. Five types of noxious gas are of potential concern. The anticipated ability of the fluidized-bed reactor to reduce each of these will be separately discussed.

1. Oxides of Nitrogen. The relatively low, uniform temperature of the fluidized-bed reactor (1300 to 1700°F) limits the formation of oxides of nitrogen. Most combustion chamber concepts require a hot, primary combustion zone to assure good combustion efficiency; it is in these hot zones that most oxides of nitrogen are formed. Because of extensive mixing of the fluidized bed, excellent combustion efficiency is realized without a hot primary zone.
2. Oxides of Sulfur. The effectiveness of limestone for control of SO_2 emission from coal combustion chambers is being demonstrated. After injection into the combustion chamber, the limestone is first calcinated to lime. The SO_2 is oxidized to SO_3 on the lime surface and then reacted to calcium sulfate ($CaSO_4$), which remains with the ash. A disadvantage of this process for coal combustion is the relatively large quantity of limestone required. Test data show that limestone may be only partially reacted because of short residence time in the furnace resulting in calcium sulfate accumulation only on the surface of the limestone. This problem is reduced with the fluidized bed since the increased residence time in the fluidized bed strongly favors the capture of sulfur by limestone. In addition, solid wastes average less than 0.5 percent sulfur and the solid waste inerts already contain significant quantities of calcium and magnesium oxides. Thus, little if any limestone additive is required when burning solid waste in a fluidized bed.
3. Hydrogen Halides. The emission of hydrogen halides, primarily HC1, can be expected to be a significant problem for future incinerators; probably more significant than SO_2 emission. Although limited experimental work exists on HC1 suppression, chemical considerations indicate that reactants similar to those previously described for SO_2 suppression may be effective for HC1 suppression.
4. Carbon Monoxide, and
5. Hydrocarbons. The highly mixed oxygen-rich environment of the fluidized-bed reactor provides very favorable conditions for complete combustion, thus minimizing carbon monoxide and hydrocarbon emission.

Finally, the fluidized bed is unique in its ability to efficiently consume low quality fuels. The relatively high inerts and moisture content of solid waste pose no serious problem and require no associated additional devices for their removal.

THE CPU-400 PILOT PLANT

The CPU-400 pilot plant development is currently nearing completion in Menlo Park, California. A systematic evolution is planned to ultimately include all major components on a pilot plant level. The planned high pressure configuration (with integrated gas turbine) consists of four primary subsystems. Three of these, broken down into their major constituent parts, are illustrated in Figure 1. These are the solid waste handling subsystem; the solid waste combustor and gas preparation subsystem; and the turbo-electric subsystem. A fourth area, the control system, is both a part of each of the other three subsystems and also a separate subsystem in itself, causing the other subsystems to interact properly with one another and respond correctly to external commands.

The purpose of the solid waste handling subsystem is to prepare the solid waste for combustion. This includes separation of the shredded material into two constituents (materials dominated by combustible and inert components, respectively), storing the combustibles until ready for use in the combustor, and metering the solid waste to the combustor. Unprocessed municipal waste is initially loaded onto the shredder conveyor by a skip loader. The conveyor modulates the feed to the shredder based upon electrical loading of the shredder motor. After shredding, the material is fed to the air classifier where the light, combustible materials are pneumatically lifted and transported to the storage bin, while the heavy, inert materials drop out for subsequent separation and recovery. Metering of the prepared solid waste fuel is accomplished through a variable speed, servo-controlled outfeed conveyor in the storage bin along with variable speed transfer and weighing conveyors to the combustor feed points.

The second subsystem of the high pressure pilot plant consists of the solid waste combustor, three particulate removal stages, ash removal equipment, and interconnecting piping and valving. The solid waste is introduced into the combustor through air-lock feeder valves. The material is burned in the solid waste combustor and the resulting hot gases are then cleaned of suspended solid material in three stages of separation. Fly ash material is removed from the particle separator collection hoppers by pneumatic transport to a baghouse filter.

The third subsystem is the turbo-electric unit consisting of a gas turbine, generator, switch gear, and load bank. The compressor section of the turbine supplies the cold air for the solid waste combustor fluidization. The resulting hot gases, after being cleaned in the separators, are used to power the compressor turbine and the power turbine. The generator is driven by the power turbine and generates power which is subsequently controlled by the switch gear. The electrical energy output of this pilot plant system will be dissipated in a combination load and light bank. In the full scale system, the electrical power will be delivered to a customer for subsequent use in the municipality.

The control subsystem interacts with these systems to control their respective outputs in response to commanded set points. This system, which features analog controllers under the supervisory control of a digital process computer, will also monitor numerous signals to provide data acquisition, logging, out-of-tolerance alarming, and status display functions.

In the low pressure configuration operated to date, the gas turbine compressor is replaced by a facility blower and exhaust gases are cooled by water spray. Consequently, there is no further discussion of the turbo-electric subsystem in this paper. Since incorporation of the process control computer into the pilot plant is incomplete at this writing, few references will be made to the control subsystem.

Solid Waste Handling Subsystem

The solid waste handling subsystem processes municipal solid waste unloaded by packer trucks at the pilot plant facility. Photographic views of the subsystem are shown in Figure 2. The flow of solid waste is illustrated in Figure 3. A 16 ft^3 skip loader is used for transfer of the raw solid waste to the shredder's conveyor hopper. Through electrical current level controls, the conveyor supplies material on demand to the shredder. Shredded material is ejected into the air classifier unit. In this unit heavy metallics, rocks, glass, etc., are dropped out; the remainder is pneumatically transported up through the chamber and into the cyclone above the storage tank where it is disengaged and deposited in the tank. The exhaust stack of the cyclone connects to a second blower on a dust filter cyclone. This unit pulls off vapors and dust from the center of the storage tank cyclone, depositing the dust in a container at floor level and venting excess air to the atmosphere.

Very little hand sorting of the delivered solid waste is conducted prior to shredding. On rare occasions an item too large for the shredder inlet (e.g., a large truck tire) is encountered; these are manually removed. Power limitations in the 75-hp shredder have dictated that massive metal items and fabric bundles (both infrequently found in municipal solid waste) also be removed. Other highly visible items such as automobile tires and mattresses, though successfully shredded on occasion, are normally removed in the interests of maintaining high through-put. Under these conditions a nominal rate of 1.5-2.0 ton/hr has been established, and over 350 tons of solid waste have been shredded and air classified through August 1972. Concessions to subscale pilot plant operation essentially disappear when 1000 hp shredders, such as planned for the CPU-400, are employed.

The shredded and air classified solid waste fuel form is a mixture whose visual appearance is homogeneous and dominated by identifiable paper products. A typical pound of the material consists of 0.30 lb of water, 0.52 lb of ash-free combustibles, and 0.18 lb of inerts (including ash). The latter two fractions are typically sub-divided as follows:

Description	**% by weight**
Paper	55
Thin metals, fine glass, and dirt	15
Wood	13
Dust	13
Plastics	2
Textiles	2

As would be expected, all of the preceding values are subject to considerable variation. As an example, moisture content in the final fuel form ranges from 10 to 40 percent by weight depending upon the origin of raw material (e.g., residential or commercial sources), time of year, weather conditions, and municipal collection policies.

Air classified solid waste fuel is shown in the storage tank in Figure 4. The accompanying view of the empty tank interior shows three of the four rotating bucket chains which sweep the floor to move material into the outfeed conveyor located in the slot. Free trailing ends of the bucket chains allow edge contact with the stored material pile regardless of pile size. Variable speed hydraulic drives in the sweep system and outfeed conveyors are controlled to maintain material levels in a small hopper at the outfeed conveyor discharge point.

The hopper volume lies above a transfer conveyor and upstream of material leveling devices that produce a fixed material height on the transfer conveyor. Fuel is delivered by the transfer conveyor to a weighing conveyor (equipped with load cells to provide continuous measurement of fuel flowrate) and then dropped through a static splitter assembly into the two airlock feeder valves.

Variable speed electric drive systems are used on the transfer and weighing conveyors. Speed command signals are both slaved to a single fuel demand signal; this implements a responsive, continuous volumetric flow element for a combustor temperature control system.

Two airlock feeder valves receive the solid waste fuel from the weighing conveyor and deliver it to the air transport lines, which in turn pneumatically deliver the material to the fluidized bed. Each valve (Figure 5) is powered by a 25-hp electric motor drive and associated gear box. As the valves turn, empty pockets in the valves receive material from the top, rotate through 180 degrees past a sealing wall, and deliver the material into the transport lines at the bottom. Material is continuously delivered to the combustor through continuous rotation of the valve.

Solid Waste Combustor and Gas Preparation Subsystem

The fluidized-bed combustor is contained within a vertically oriented cylindrical pressure shell with dished heads. The outside diameter is 9.5 feet and overall height is 23.5 feet. Three layers of insulation protect the 3/8-in. carbon steel pressure shell cylindrical sections from high combustion zone temperatures. A wear-resistant firebrick inner liner is backed up by a liner of insulating brick. These refractory layers are separated from the shell by a thin layer of packed ceramic fiber insulation designed to isolate the shell from stresses induced by differential thermal expansion. Insulation in the top dome is provided by a castable refractory held in place by standard hangers.

The fluidized bed is supported by a flat carbon steel plate welded to the pressure shell and covered by two layers of castable refractories that provide insulation and wear resistance. Penetrating this assembly are 161 2-in. pipes capped with wire mesh air diffusers. Other penetrations from the air plenum chamber beneath the plate permit bed temperature and pressure measurements.

The circular cross section fluidized bed has an area of 40 ft^2 and is designed to operate with a superficial velocity in the 5 to 7 ft/sec range. A nominal 2-ft bed (unfluidized state) is used together with a 12-ft freeboard (unfluidized bed surface to exhaust duct centerline).

Penetrations through the pressure shell and refractory insulation into the fluidized bed provide for two solid waste feedpoints. Two feedpipes bolted to outer shell bosses extend into the bed. Solid waste is fed into the bed along the length of these pipes via a slanted cut on the bottom side. The design and positioning of these pipes is based on earlier tests where oxygen concentration measurements established the dimensional characteristics of combustion zones. The result is a configuration which, in low pressure testing, has demonstrated very satisfactory operation with respect to geysering due to feedpipe air flow, minimization of local fuel-rich zones, and reduction of heat release above the bed.

Other bed penetrations provide for possible removal of excess bed material and for six oil guns to permit fluidization combustion of auxiliary diesel oil. This normally unused auxiliary fuel, available primarily as a developmental tool for backup service in maintaining or establishing desired test conditions, is mixed with air and carried through the inner of two concentric pipes. The outer pipe of each gun carries cooling air.

Initial bed heating is accomplished by hot products of combustion from an oil burner located in the top dome of the combustor. This downward firing burner forces hot gases through the bed in a "back heating" mode that is capable of heating the bed from ambient conditions to 1100°F in 90 minutes. This bed temperature, being above the auto-ignition temperature of either solid waste or diesel oil, is an appropriate initial condition for successful fluidized combustion.

In the low pressure configuration, fluidizing air is supplied to the combustor's air plenum by a 125-hp positive displacement blower which can deliver up to 7000 scfm at 3 psig. The blower also supplies fuel transport air in a parallel path to the fluidized bed. By appropriate valving, the same blower is used to drive the back heating mode.

The top cylindrical section is removable and contains the exhaust port, instrumentation, and observation ports. Exhaust into the first particulate removal stage is carried by a double-walled pipe with 26-in. carbon steel outer wall and 20-in. type 310 stainless steel inner liner. The annular space is packed with ceramic fiber insulation.

Two particulate removal stages have been tested in low pressure operations by the time of this writing. The first, known as the alumina/sand separator, treats a problem peculiar to fluidized-bed combustion of solid waste fuel; the removal of inert particles of elutriated bed material, and the handling of particles generated by the presence of aluminum in the fuel. While the majority of aluminum is removed by air classification, about 0.25 percent by weight of the processed fuel is aluminum in the form of foils, beverage can pull-rings, and other thin or trapped particles. The fluidized bed melts, fragments, and partially oxidizes this material into particles having molten internal aluminum and a frozen oxide (alumina) surface shell that inhibits further rapid oxidation. Previous testing has shown that elutriated particles of this type tend to generate sizable deposits with high aluminum content on impingement surfaces in the exhaust gas stream. The possible participation of soft (at combustion temperatures) bottle glass particles or other binding agents in this deposition mechanism should not be discounted, even though present evidence does not seem to indicate that they play a dominant role.

A promising solution to the problem of handling partially molten aluminum and other sticky particulates is to provide a curved surface in the first separator stage where the turning exhaust gas can produce a controlled deposit. Scrubbing action associated with the continual impingement and turning of particle laden exhaust gases acts to promote further oxidation at the deposit surface. It also promotes the necessary erosion of resultant alumina-rich particles to stabilize deposit shape and size. Most of the inert alumina-based particles thus generated as well as the silica-based elutriation particles are then collected in the settling chamber formed by the bottom of the vessel. In testing to date, these separated particles have been allowed to remain in the settling chamber. For future tests, residue particles collected in the alumina/sand separator stage will be removed on a continuous basis.

Fly ash and fine bed material particles are removed in two stages of inertial separation. The first inertial separator stage has been successfully tested by the time of this writing and the final stage (similar design features) will be added in the immediate future. An inertial separator assembly consists of an inertial separator tube holder with a residue collecting hopper, an insulated cylindrical housing with inlet and outlet flanges, a dished, insulated flanged head, and stainless steel liners. As shown in Figure 6, the hot gases enter the inlet cavity to the cyclone tubes through an internal circumferential passage. The entering gas then turns and flows through the spirally finned annular section of each tube. These fins impart rotational motion into the gas which centrifuges the particles to the outside wall. Particles spinning along the outside wall of the tube will decelerate and fall through the opening at the bottom of the tube into the collecting hopper. To avoid plugging, the gravitational flow of particles from each tube is assisted by a secondary bleed flow. Particulates are removed from the hopper through the opening on the bottom of the hopper cone. Gas which enters the cyclone tubes turns 180 degrees and exits through the center tube into the outlet manifold of the vessel. In the first inertial separator, space is provided for 48 6-in. tubes; half of the spaces are plugged for low pressure operation, however. When incorporated, the second inertial separator will have space for 100 3-1/2-in. tubes.

Material from the inertial separator hopper is pneumatically transported to the baghouse filter through a finned line. The high temperature baghouse assembly includes a puffback bag cleaning system, an exhaust blower, a holding bin, and unloading valve.

A photographic view of current subsystem components (Figure 7) also shows the gas turbine in the foreground. Present plans are to install and integrate this turbine into the pilot plant by the end of 1972.

SUMMARY OF COMBUSTION TEST RESULTS

The seventh combustion test conducted on the low pressure configuration described in preceding paragraphs was completed in August 1972. The test featured 35 hours of fluidized combustion and produced an extensive quantity of data while consuming over 69,000 lb of shredded, air classified solid waste. During one particular 24-hr period of special performance interest, a number of solid waste and exhaust gas stack samples were drawn for laboratory analysis. Most of the discussion to follow will be based on data obtained in this latter period.

A laboratory facility has been established at Combustion Power Company to perform many of the required experiments on material samples drawn from the pilot plant process operation. Included in the installation are equipment and procedures for the determination of:

1. Solid waste moisture fraction,
2. Solid waste inerts fraction,
3. Solid waste heating value,
4. Granular material size distributions,
5. Particulate loading and size distributions from gas samples, and
6. HCl gas concentrations in exhaust gas samples using titration techniques.

Fuel Properties of Prepared Solid Waste

A combination of 12 laboratory samples and adjustments based on long term pilot plant mass balance measurements yielded the following average weight distributions for the shredded and air-classified solid waste used during the 24-hr period.

Ash-free combustibles	0.516
Moisture	0.301
Inerts	0.183

A series of 12 bomb calorimeter experiments in the laboratory on dried parallel samples produced an average higher heating value of 6437 Btu/lb. Using an approximate ultimate analysis of $C_{30}H_{48}O_{19}$ for the combustibles fraction and converting to a lower heating value based on the combustibles only, a corresponding average value of 8087 Btu/lb of combustibles is found. This result is in very good agreement with values determined by applying heat balance relations to observed pilot plant temperature and flow measurements. It also correlates well with expected values for a cellulose-like material such as approximated by the $C_{30}H_{48}O_{19}$ formulation.

Compared to more conventional fuels, the heating value of solid waste is in a sense degraded by the presence of inerts and moisture in greater-than-normal concentrations. On the other hand, the greater mass flows of combustibles and water vapor required to vaporize and heat the water are exploited by the gas turbine cycle as a natural form of water injection. In addition, fluidized beds have a demonstrated ability to consume "low quality" fuels and hence to eliminate any need for a drier or other additional fuel preprocessing.

With the preceding fuel properties, the steady state combustor operating point described in Table 1 is typical. Note the 178 percent excess air level associated with this operating condition, which is one of the reasons that very high combustion efficiencies are realized.

Table 1. TYPICAL OPERATING POINT OF THE LOW PRESSURE CPU-400 PILOT PLANT CONFIGURATION

Solid waste fuel flowrate, lb/min	36.6
Combustor air flowrate (includes feed line), lb/min	330
Excess combustor air, %	178
Solid waste fuel inlet temperature, °F	70
Air inlet temperature, °F	130
Exhaust gas exit temperature, °F	1,500
Total combustor heat release, Btu/min	152,700
Heat loss to ambient (two vessels), Btu/min	5,000
Mole fraction of major exhaust gas contituents:	
Oxygen	0.122
Carbon dioxide	0.063
Water	0.099
Inert mix of nitrogen and argon	0.716

The excess oxygen ratio is strongly influenced by fuel moisture fraction. If moisture fraction is increased to 0.50, for example, and inerts fraction is reduced to 0.131 (as would happen if the added moisture was obtained by direct addition of water to the previous fuel mixture), then the required solid waste flowrate jumps to 64.9 lb/min, and the excess oxygen percentage drops to 119 percent. Further increases in moisture fraction produce even sharper drops in excess oxygen ratio so that it is not possible to use moisture fractions much over 0.60. An interesting interpretation of the preceding numbers is that the combined injection of 18.5 lb/min of water and 30 percent moist solid waste raises the consumption of the latter (and hence the total heat release) by 27 percent to 46.4 lb/min.

Fluidized-Bed Combustor Performance

With a large, relatively shallow fluidized bed served by one or two feed pipes, a small but significant portion of the total combustion process occurs in the lean phase or freeboard above the dense bed. For a 40 ft^2 bed area 2 feet deep, for example, bed to exhaust temperature rises of 260° and 160°F were observed with one and two feed pipes in operation, respectively. Since these levels of gas phase combustion (afterburning) are quite stable for well controlled solid waste injection, it has been concluded that satisfactory operation can be obtained in either case at low pressure conditions.

Very high overall combustion efficiencies have been demonstrated by several approaches. First, careful heat balance calculations based on process measurements have consistently produced apparent heating values that, when compared to laboratory calorimeter results, produce efficiency values slightly in excess of 100 percent. Flow measurement errors and occasional high heating value components in the solid waste not found in laboratory samples are the probable explanations. A second method based on a carbon balance as indicated by continuous exhaust gas measurement of CO_2, CO, and hydrocarbons plus post-test analysis of free carbon in the separator residue stream, generated an average efficiency value of 99.76 percent. Finally, there is no visible exhaust smoke, odor, or other common evidence of an inefficient process.

System Inert Material Balance

After installation of the combustor was completed it was loaded with a 2-ft "starter" bed of commercial 16-mesh beach sand having a bulk density of about 100 lb/ft^3. After 18 hours of fluidized operation on solid waste fuel in the first six tests, the cooled bed was observed to still be 24-in. deep and free from excessively large particles or clinkers. No material had been removed except through the elutriation process. The size distribution was somewhat larger (i.e., more fines and more coarse particles) and the bulk density had dropped to 91 lb/ft^3.

The bed generated in this fashion was successfully used in the following 35 hr test. Again, no material was removed except by elutriation. Post-test analysis and inspection then showed that the bed was 25-in. deep and had an average bulk density of 87.5 lb/ft^3. Thus the 7,300-lb bed processed and elutriated more than 12,300 lb of inerts in 35 hours while only growing about 10 lb. This natural replenishment of bed material by the inert content of the fuel is a phenomenon peculiar to solid waste combustion. Earlier long duration tests (240 hours) on a 2.2 ft^2 bed coupled with computer simulation of transient size distribution histories had shown that the steady state bed could be expected to have quite acceptable properties. Consequently, the bed maintenance and elutriation control problems appear to be grossly simplified.

In the 35-hour test, 72.0 percent of the elutriated material (i.e., 8890 lb) was collected in the settling chamber of the alumina/sand separator vessel. This inert granular residue has considerable promise as a construction material. Another 25.4 percent (3140 lb) was collected by the first stage inertial separator in the form of fine fly ash. The remaining 2.6 percent, 320 lb of very fine fly ash, left with exhaust gases. Most of this latter material will be collected by the second stage inertial separator in subsequent tests.

Gas samples were withdrawn from the first stage inertial separator outlet at 3-hour intervals for laboratory analysis of particulate loading. Early samples showed a total loading of 0.057 gr/scf and a loading of 0.025 gr/scf for particles greater than 5 μm in size; both values are indicative of satisfactory first stage performance. Later markedly higher values confirmed post-test findings that the hopper beneath the 24 active tubes has failed to drain properly and therefore progressively plugged the ash discharge sections of some tubes, rendering them ineffective. Twin vibrators have been installed to solve this problem in future tests.

Exhaust Gas Composition

A set of instruments has been installed for on-line concentration measurements of six specific constituents of the exhaust gas. A seventh gas chromatograph instrument for the on-line measurement of HCl is under development and will be added to replace current laboratory titration procedures. The gas sampling, conditioning, and distribution systems are integrated with the instruments and analog recorders in a mobile, rack mounted, complex. The six current instruments, measuring principles, and associated gases are:

1. Beckman 715, Polarography, Oxygen;
2. Beckman 315B IR, Infrared, Carbon dioxide;
3. Beckman 400, Flame Ionization Detection, Hydrocarbons;
4. Beckman 315B IR, Infrared, Carbon monoxide;
5. Theta LS-800AS, Electrochemical reaction, Sulfur dioxide; and
6. Theta LS-800AN, Electrochemical reaction, Nitrogen oxides.

The sampling probe leads into a stainless steel sampling train that includes particle removal elements, gas cooling and drying, controlled reheating to 100°F, a common manifold, and flow control elements for each instrument. Various calibration and zero adjustment methods have also been incorporated.

Measurements from the recent pilot plant test are presented in Table 2 together with pertinent emission standards. All instrument records were relatively steady and free from apparent anomalies considering the potential heterogenous composition of the fuel form. The 12 discrete laboratory samples (2-hr intervals) and subsequent HCl analyses showed more variance from a low value of 40.5 ppm to a high of 122.4 ppm.

The first two entries of Table 2 correlate well with analytical predictions such as contained in Table 1. The very low values for the second two, coupled with the average 45.8 ppm of free carbon (weight basis), are indicative of the high combustion efficiency. Sulfur dioxide presently appears not to present a pollution control problem, probably owing to the relatively low sulfur content of municipal solid waste and the apparent capture of existing SO_2 by the bed material.

Measured NO_x levels are much closer to proposed standards and somewhat higher than originally expected. As confirmed by other investigators, much of the nitrogen

Table 2. EXHAUST GAS CONSTITUENTS AND REFERENCE DATA

Constituent	Proposed or expected EPA emission standards (maximum)	Mole fractions measured during 24-hour CPU-400 pilot plant test
Oxygen	-	11.7 %
Carbon dioxide	-	6.8 %
Hydrocarbons	800 ppm	14.4 ppm
Carbon monoxide	2000 ppm	41.7 ppm
Sulfur dioxide	300 ppm	20 ppm
Nitrogen oxides	NA[a]	162 ppm
Hydrogen chloride	NA	90.3 ppm

[a] Not available.

emitted in these compounds derives from the fuel rather than the combustion air. Other experience, however, indicates that the concentrations may be expected to drop as pressure is increased.

Depending on emission standards yet to be established, the measured levels of HCl pose a potential control requirement. There appears to be a number of effective remedial measures that rely upon fluidized-bed characteristics, however, if forthcoming pressurized combustor tests should firmly establish a requirement.

CONCLUSIONS

Shredding and air classification operations on municipal solid waste produce a fuel form having very satisfactory physical and chemical properties for energy recovery through combustion in a fluidized-bed reactor. A solid waste handling subsystem with reliable components has been developed and extensively operated.

The fluidized-bed combustor has fulfilled its promise as a highly efficient, easily fed, readily controlled reactor of simple design and capable of utilizing low quality fuels.

Grade 16 silica beach sand is an acceptable starter bed material. The inert content of typical solid waste provides a natural bed make-up material that leads to a satisfactory steady state bed composition. As a result, the need for elaborate bed maintenance or anti-elutriation devices is minimized if not eliminated.

Test results to date show no problem with fluidized-bed residue buildup. Relatively large metal and inert particles entering the active bed experience gradual oxidation or attrition to typical bed size particles and eventually are elutriated to be collected by particle separators. No bed material agglomeration is experienced in the 1270 to 1450°F bed temperature range.

Gas phase combustion above the fluidized bed has been reduced to acceptable values without resorting to undesirable remedies such as extensive internal bed structures, numerous fuel feed points, deeper bed, multi-stage combustors, etc.

Earlier combustor freeboard and exhaust system deposit problems due to the aluminum content of solid waste appear to have been solved.

Performance of the first stage inertial separator has been very promising and is expected to further improve at high pressure conditions. Reliable handling and transportation of removed hot, fine fly ash has posed development problems.

Exhaust gas sampling instruments indicate that the pollution control effort required for the CPU-400 process can be expected to be minimal.

No evidence of serious hot gas subsystem material corrosion or erosion problems has been found.

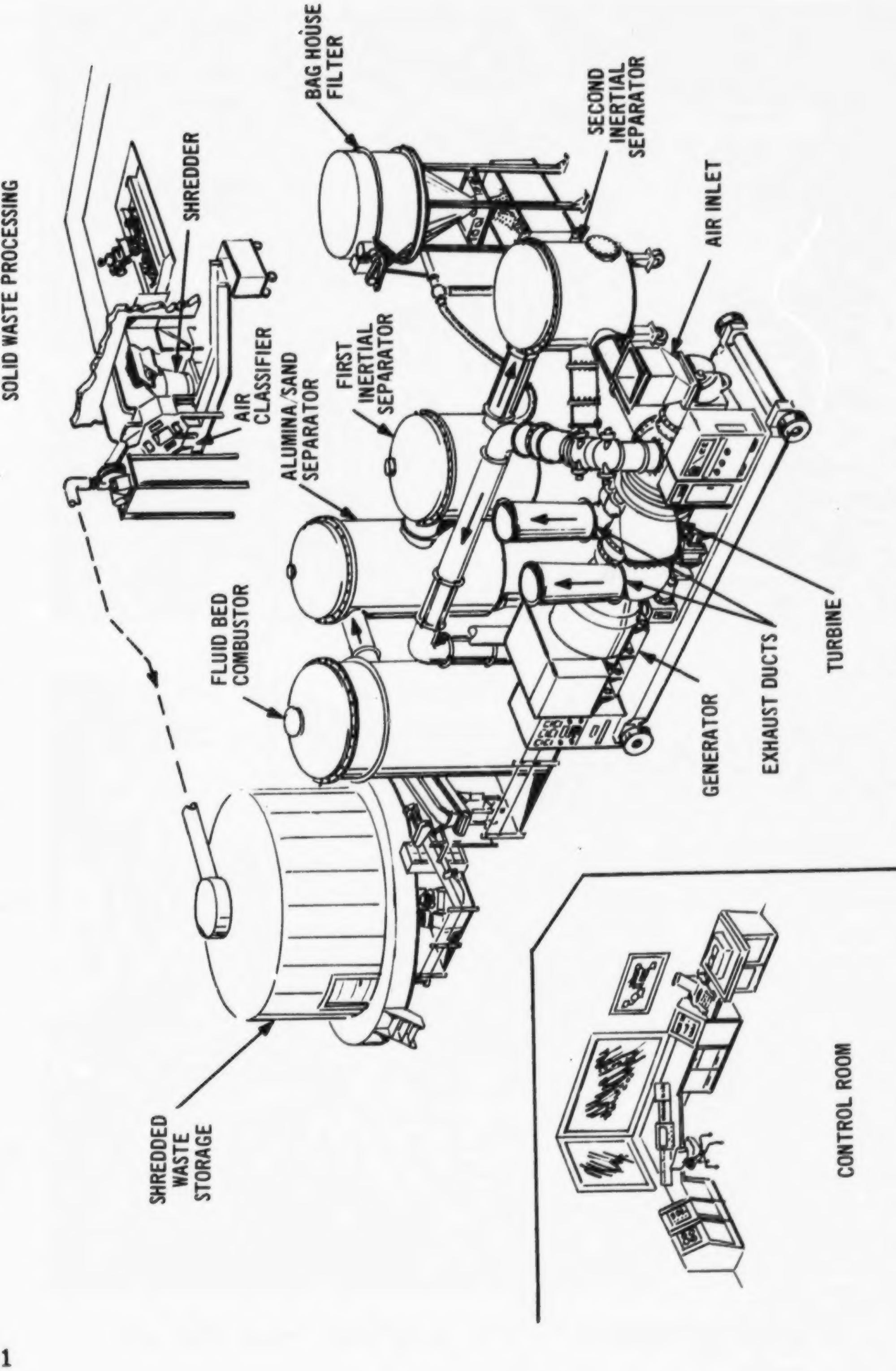


Figure 1. CPU-400 pilot plant pictorial.

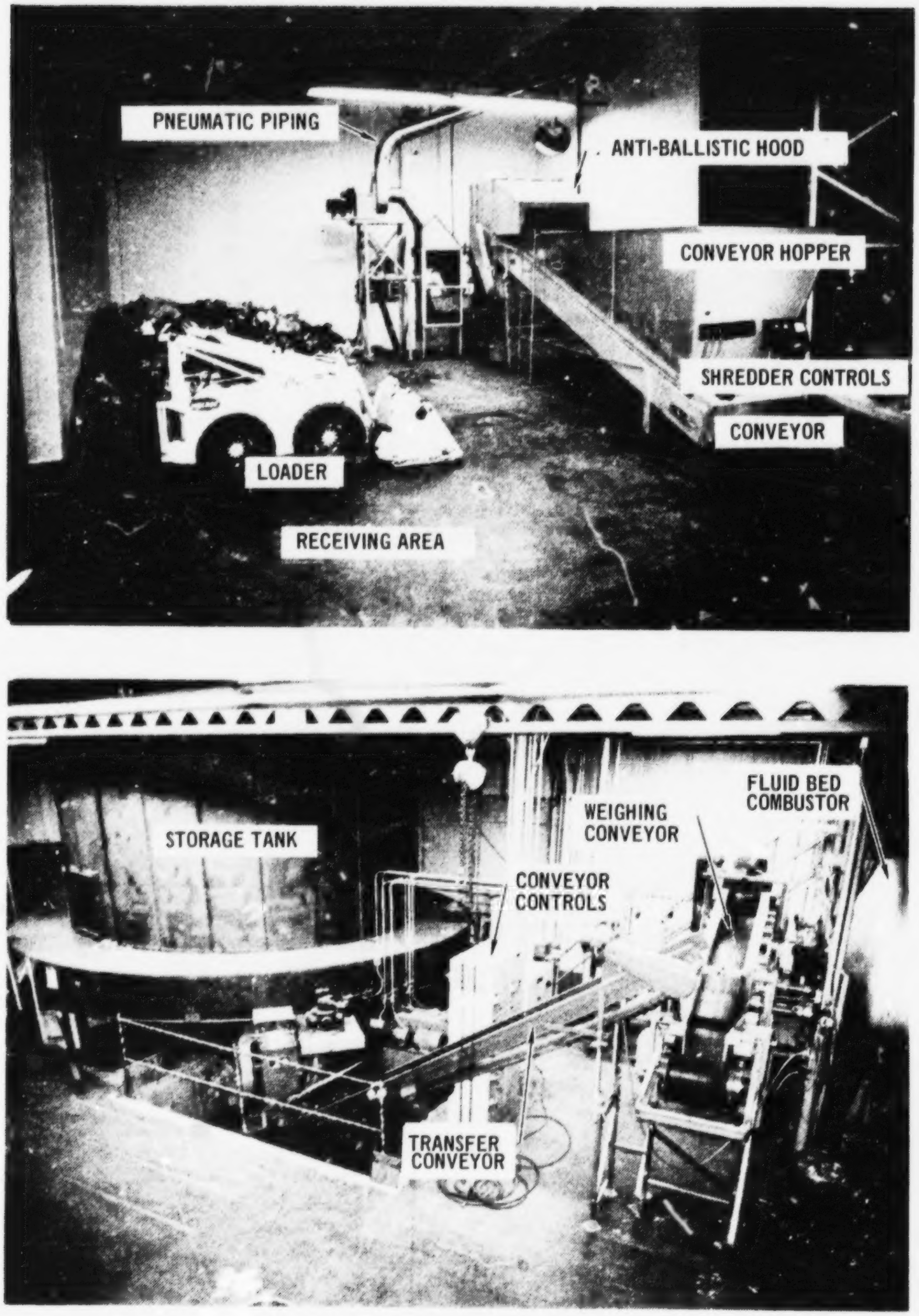


Figure 2. Photographic views of the solid waste handling subsystem.

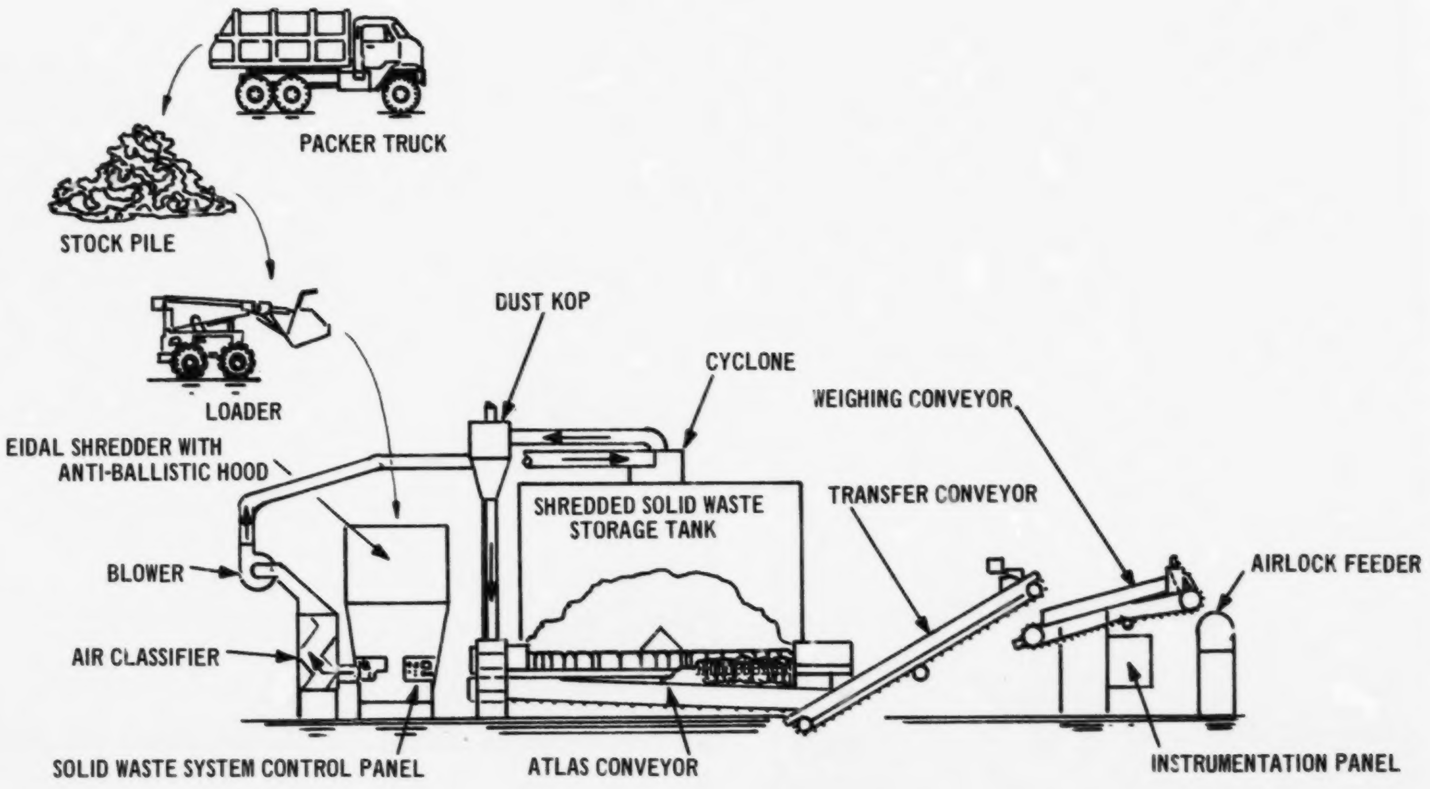
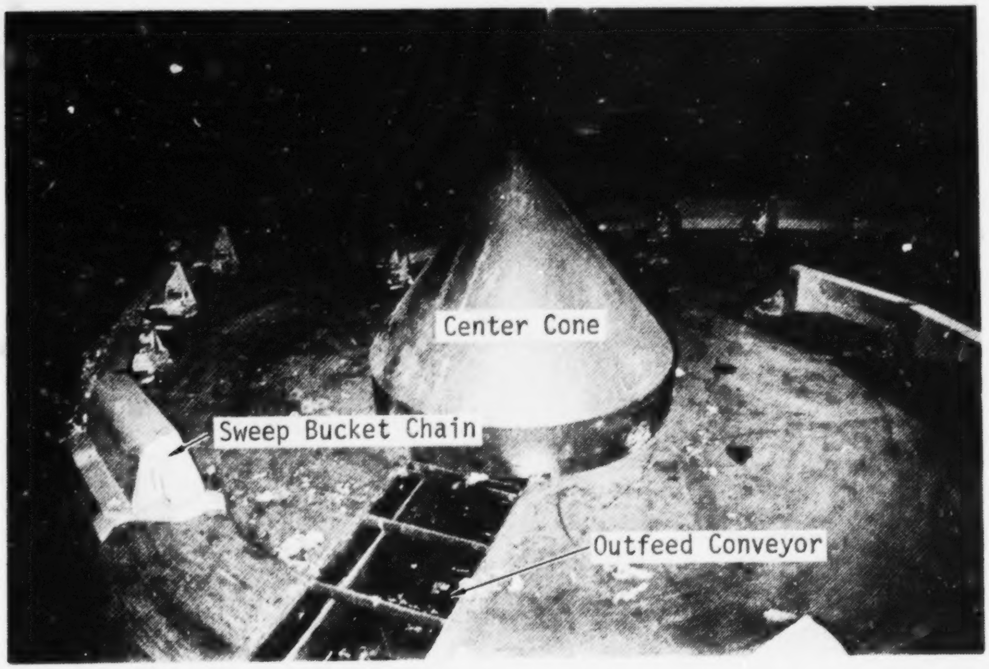


Figure 3. Pilot plant solid waste handling subsystem schematic.



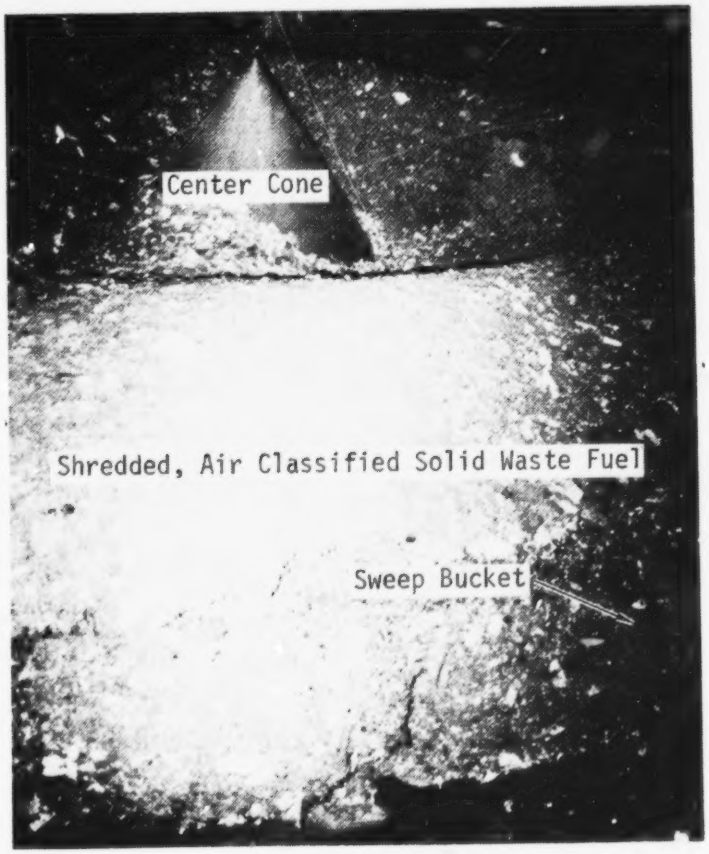


Figure 4. Photographic views of solid waste storage tank interior.

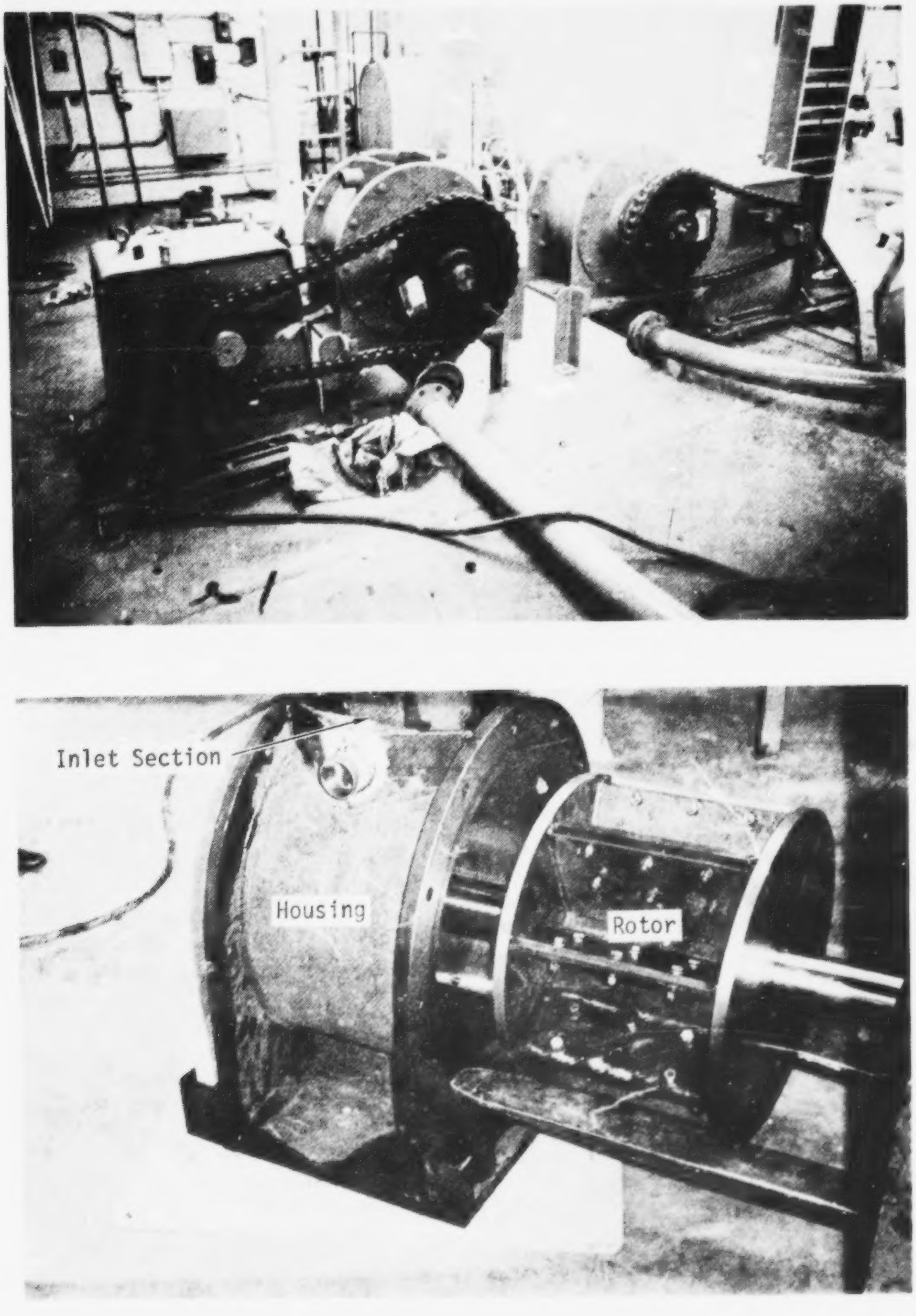


Figure 5. Airlock feeder valve.

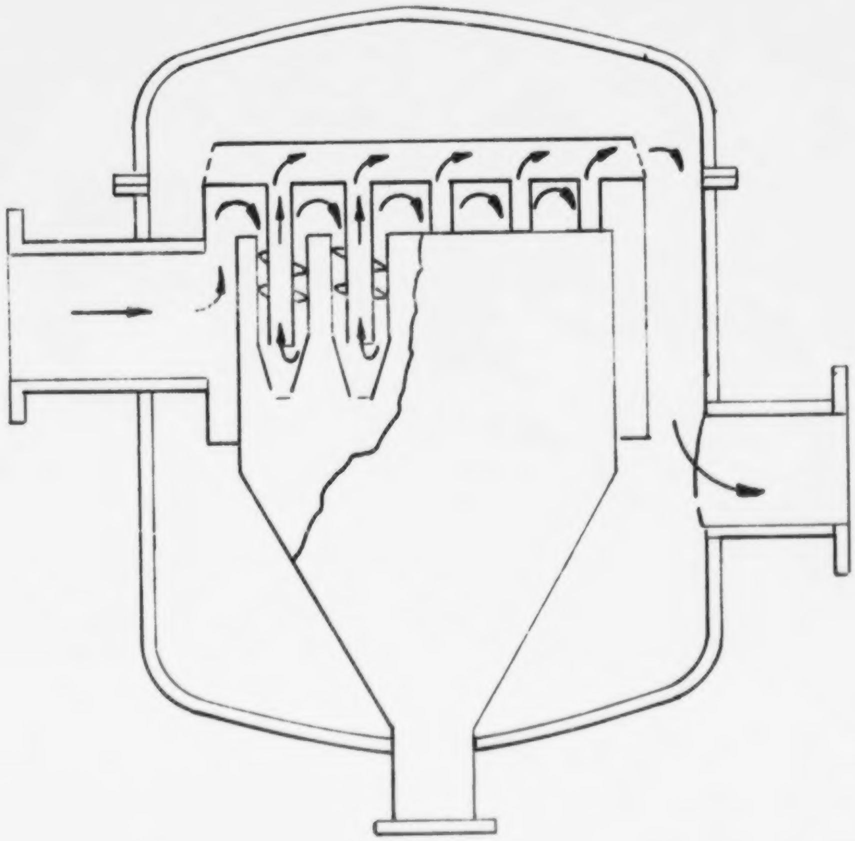


Figure 6. Inertial separator schematic.

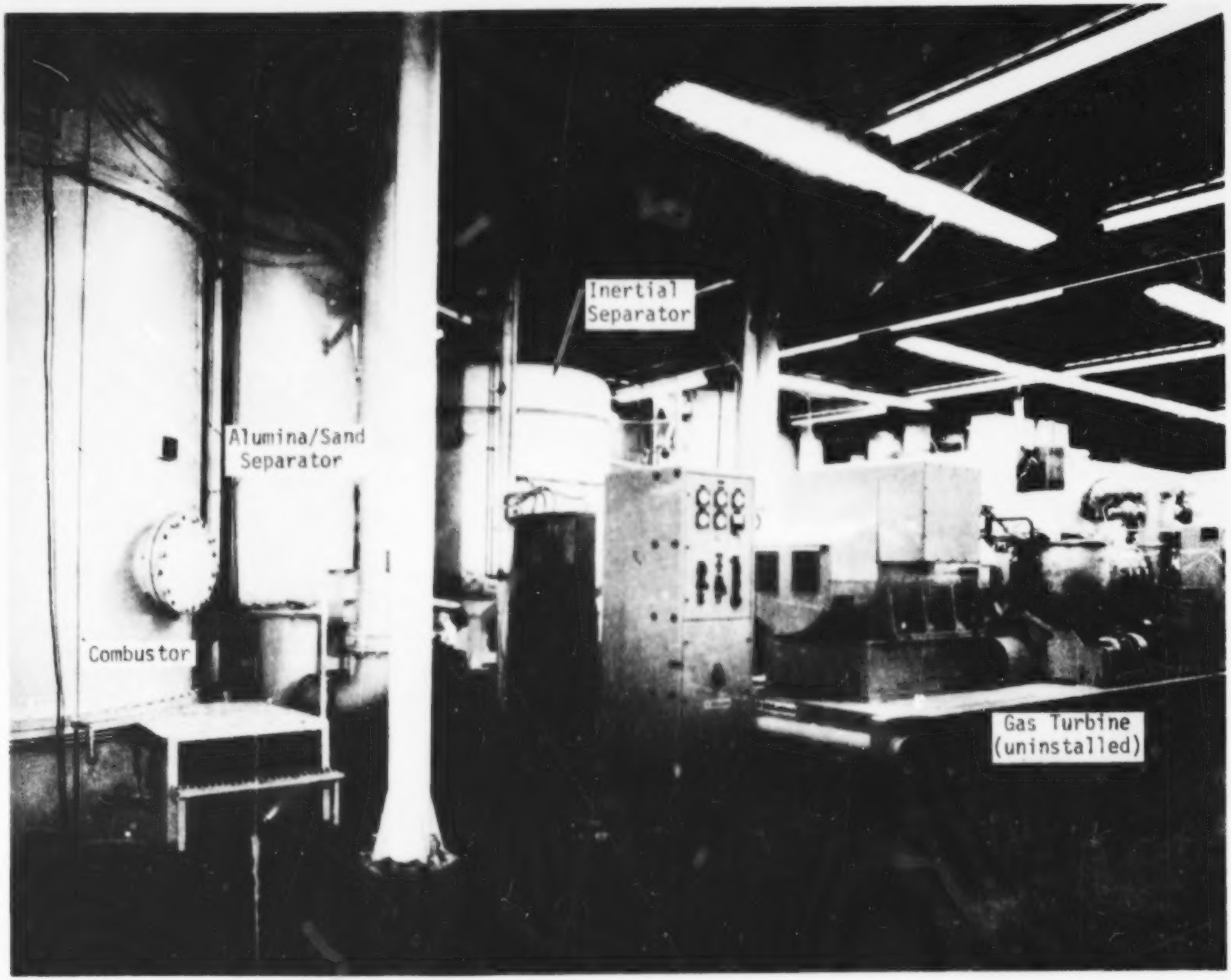


Figure 7. View of solid waste combustor and gas preparation subsystem.

4. FLUIDIZED-BED COMBUSTORS USED IN HTGR FUEL REPROCESSING

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ABSTRACT

High-temperature gas-cooled reactors (HTGR) utilize graphite-base fuels. Fluidized-bed combustors are being employed successfully in the experimental reprocessing of these fuels. This paper presents a general discussion of the reprocessing method and describes the two types of fluidized beds being used.

INTRODUCTION

The high-temperature gas-cooled reactor (HTGR), as developed at Gulf General Atomic, is a helium-cooled, graphite-moderated reactor. The fuel in an HTGR consists of fissile microsphere particles containing U-235, recycle microsphere particles containing U-233, and thorium fertile particles contained in a hexagonal fuel element, shown in Figure 1. The HTGR fuel recycle operation consists of shipping spent fuel to recycle facility, reprocessing the fuel to recover the U-233 and U-235, refabricating the U-233 and U-235 into recycle fuel, shipping the refabricated fuel from the recycle facility to the reactor, and ultimately storing the radioactive fission product wastes.

The fuel reprocessing sequence starts with the head-end operation shown in Figure 2, in which the fuel in the HTGR fuel element is separated from the graphite body by crushing and fluidized-bed burning. Subsequent head-end operations separate particles containing U-235 from ash containing U-233, thorium, and fission products. The metal oxide ash is dissolved to create a solution of uranium, thorium, and fission products; the silicon-carbide-coated U-235 is the residue. The U-235 is separated mechanically, and the uranium and thorium are recovered individually from the fission products. The recovered U-233 and thorium are stored for reuse as fuel. The radioactive wastes are disposed of in appropriate storage facilities.

HEAD-END REPROCESSING

Head-end reprocessing for HTGR fuel consists of a crush-burn-leach process.[1] Fuel element size reduction is the first step in head-end reprocessing. Two major criteria govern this step: (1) the fuel must be crushed to a suitable size for maintaining fluidization quality in the fluidized-bed burners, and (2) the crushing system must minimize fuel particle breakage to prevent undesirable crossover of fissile and fertile product uranium.[2]

A three-stage crushing system has been adopted for the reprocessing plant, based on the experimental testing of commercially available equipment using full-sized fuel elements. This crushing system is presently being tested.

Primary reduction is done in a large, overhead eccentric jaw crusher; secondary reduction in a small, overhead eccentric jaw crusher; and tertiary crushing in a double-roll crusher. The tertiary crusher product, nominally minus 3/16 in., is pneumatically conveyed to the fluidized-bed burner feed hoppers.

Crushed fuel is fed to the top or base of a continuous, exothermic fluidized-bed burner, shown in Figure 3, by an auger feeder. (The term exothermic is used to describe the burner that generates sufficient heat to maintain operating temperature.) The feed rate is automatically controlled by the off-gas carbon-monoxide concentration, which has been shown to be proportional to the graphite surface area exposed in the bed.

Both the crushed graphite and the silicon-carbide-coated fissile particles serve as the fluidizing media. The heat generated by burning is removed by forced-air cooling in a clamshell jacket surrounding the burner and an off-gas heat exchanger. The fluidizing gas fed to the burner is oxygen with a small amount of inert gas (i.e., CO_2, N_2), and the flow is automatically controlled to maintain the bed temperature. The burner product removal rate is automatically controlled by the bed pressure drop, which is proportional to the bed weight.

The burner off-gas with entrained fines is passed through a cyclone separator and a sintered metal filter for fines removal, before being cooled and proceeding to off-gas treatment. Off-gas treatment removes the fission products including noble gases before release to the environment. Fines are presently being recycled to the burner in the experimental program.

If the exothermic burner is operated with top feed and no fines recycle, the elutriated fines from the burner (both TRISO/TRISO and TRISO/BISO flowsheets) are also added to the feed stream for the last burning step. This mixture now constitutes the feed material for the endothermic (requiring heat input from a furnace to maintain operating temperature) fluidized-bed burner.

The exothermic burner product is fed to a batch-operated endothermic fluidized-bed burner, shown in Figure 4, where the remaining graphite is burned and the thorium and uranium-oxide kernels are exposed. The silicon-carbide-coated fissile particles serve as the inert fluidizing media. The feed stream to the endothermic burner will not sustain exothermic burning to the low carbon level required in the subsequent processing steps. The burning in the endothermic burner, therefore, proceeds from exothermic conditions, with the heat removed from a clamshell surrounding the burner, to endothermic conditions, with heat supplied by resistance heaters located in the clamshell. The off-gas from the burner is treated in the same manner as that from the exothermic burner. The product from the endothermic burner is pneumatically conveyed to the leaching system.

The thorium and uranium oxides are dissolved in acid thorex [13M HNO_3 - 0.05M HF - 0.01M A1 $(NO_3)_3$] in a steam-jacketed cylindrical vessel with gas sparge mixing. This leaching vessel is run as a refluxing, batch leacher.

The insoluble silicon-carbide-coated fissile particles and the unburned carbon must be separated from the mother liquor before the solution can be fed to the solvent extraction system for uranium purification and thorium recovery. A centrifugal separator receives the entire slurry from the leacher. Solids retained on the centrifuge screen are washed with fresh leach solution, which becomes the leach solution for the next batch of solids from the endothermic burner. The washed solids from the leacher are then air-dried and transferred to a screen classifier, where the fissile particles are separated from the silicon-carbide hulls. The waste solids are processed as wastes, and the fissile particles are stored for later processing, by a method similar to that for the fertile particles, to recover and purify the uranium.

The clarified leach solution is evaporated and steam-stripped to an acid-deficient condition for use as feed to an acid thorex[3,4] extraction process. The acid thorex solvent extraction process is used for the decontamination and purification of the U-233 and thorium and for the separation of the U-233 and thorium from each other.

FLUIDIZED-BED BURNERS

Exothermic Fluidized-Bed Burners

Figure 3 depicts the exothermic fluidized-bed burner presently being used in the experimental program. Exothermic burners with both 4-in. and 8-in. diameters are being used; construction of a larger burner is planned for early next year. The large burner will become the full-sized commercial plant test unit. Preliminary nuclear criticality calculations have shown that this burner can be about 16 inches in diameter.

Operability

The exothermic burners have been operated on a routine basis for the last 18 months. Startup is initiated by heating a charge of coke (1200 g for the 8-in. and 400 g for the 4-in.) to ignition temperature (700°C) with a carbon monoxide-oxygen gas mixture. This gas mixture is introduced into the burner with a standard cutting torch that is ignited by two spark plugs. After the coke is ignited, fluidizing oxygen and the graphite-base feed are introduced.

The steady-state bed properties of the exothermic burners are listed in Table 1; the bed contains 2 to 5 percent burnable carbon.

Table 1. AVERAGE BED PROPERTIES FOR EXOTHERMIC BURNERS[a]

Size	Wt % of total sample
-3/16 in. + 1/8 in.	0
-1/8 in. + 869 μm	0.2
-869 μm + 550 μm	67.9
-550 μm + 420 μm	4.9
-420 μm + 375 μm	0.2
-375 μm + 250 μm	1.0
-250 μm + 125 μm	5.5
-125 μm + 74 μm	4.3
-74 μm + 44 μm	2.8
-44 μm	13.2

[a] Burnable Carbon: 3.0%
Average Particle Size: 590 μm

Steady-state operation is achieved for experimental purposes in about 4 hours by adding the estimated steady-state bed composition directly to the burner immediately after startup. Product removed after 4 hours is also near steady-state and exhibits about the same properties as the bed. The present feeding method is a variable-speed auger controlled automatically by the carbon-monoxide concentration in the off-gas. The nominal off-gas concentrations are 1 to 3 percent carbon monoxide, 0 to 6 percent oxygen, and 60 to 95 percent carbon dioxide. This composition is a function of the inert fluidizing gas diluent (i.e., air versus CO_2). Product removal rate is automatically controlled using the pressure drop across the bed and regulating a variable-speed drive motor on the product removal auger. Bed temperature is automatically controlled by regulating the fluidizing oxygen supply to the bed. Temperature profiles, off-gas compositions, bed and filter pressure drops, and mass flows of important streams are monitored continuously.

Fluidization quality has been difficult to define. Normal operation is with a well-mixed bed that occasionally slugs. Distributor plates are not presently being used but will be fully tested in the near future. At present, operation is with a cone base and a ball check valve. Preliminary tests of perforated plates, bubble caps, and sintered metal screens were all successful to some extent, and the beds appeared to maintain good fluidization quality.

Feed

The feed to the burners is presently defined as minus 3/16-in. graphite-based material. This feed size was established by gradually increasing the size from minus 1/16 inches to minus 1/4 inches. Poor fluidization occurred with minus 1/4-in. feed, as witnessed by local "hot spots" in the bed. Returning to minus 3/16-in. feed eliminated this problem. Table 2 shows the average properties of the exothermic burner feed.

Table 2. AVERAGE PROPERTIES OF EXOTHERMIC BURNER FEED[a]

Size	Wt % of total sample	% burnable carbon in fraction
-3/16 in. + 869 μm	64.6	100
-869 μm + 550 μm	25.0	21
-550 μm + 420 μm	1.5	78
-420 μm + 375 μm	1.5	97
-375 μm + 250 μm	1.5	98
-250 μm	5.9	98

[a] Top Density: 1.25 g/cm^3
Bulk Density: 1.08 g/cm^3
Angle of Repose: 35°
Average Burnable Carbon: 80%
Average Particle Size: 854 μm

Heat transfer

The heat transfer problem encountered is somewhat different from that occurring in most fluidized-bed work; because of nuclear criticality considerations, the cooling medium is limited to air. Figure 5 shows the heat balance for a typical 8-in. diameter exothermic burner run. The over-all heat transfer coefficients for the off-gas heat exchanger and clamshell cooler are also shown.

Fines recycle

One of the major problem areas in exothermic burner operation is the burning of fines that have elutriated from the burner. Since burning efficiency and radioactive hot-cell constraints play a major role in the process, work has focused on burning the fines by recycle to the bed rather than in a separate fines burnup cell.

Fines carryover has been about 22 percent of the burn rate when the furnace has operated at normal conditions; i.e., at a burn rate of approximately 200 g/min and a superficial fluidization velocity of 3 to 4 ft/sec. Of these fines, about 98 percent is collected by a cyclone and 2 percent by a filter chamber. The elutriated fines are described in Figure 6.

The present operating mode is to recycle the fines by blending them with the graphite-base feed stream. This composite is fed to the bottom of the fluized bed, and the fines appear to burn successfully when steady-state is achieved.

The nominal burn rate for the 4-in. and 8-in. exothermic burners is about 33 g carbon/hr-ft^2, which corresponds to 50 and 200 g carbon/min, respectively. A burn rate of 50 g carbon/hr-ft^2 (corresponding to 75 and 300 g carbon/min, respectively) is planned for the 4-in. and 8-in. exothermic burners. Burn rates of 125 and 475 g carbon/min for the 4-in. and 8-in. burners (about 84 g carbon/hr-ft^2) have been achieved for short periods. Operation at these high burn rates is not possible for long periods because of limitations with existing equipment. Future studies will be aimed at defining these values over long-term run conditions.

Scale-up

The design considerations for the larger exothermic fluidized-bed burner include both a theoretical approach and scale-up factors from the 4-in. and 8-in. burners. To date, the problems encountered for scale-up have been in defining both a suitable transport disengaging height and exact heat transfer values. Although the theoretical and experimental predictions of heat transfer values in the area of the bed-wall-clamshell are in agreement, it is difficult to define the proper heat transfer coefficient in the transport disengaging height in which the overall heat transfer coefficient rapidly decreases with reactor height. These values will be determined experimentally in future experiments in which "sectioned" clamshell coolers will be utilized.

Endothermic Fluidized-Bed Burners

Figure 4 depicts the 4-in. diameter endothermic fluidized-bed burner presently being used in the expermental program. It is planned to convert the 8-in. diameter exothermic burner to an endothermic burner by adding resistance heaters to the clamshell interior and moving the filter chamber to directly above the burner for future scale-up testing.

Operability

The endothermic fluidized-bed burner has been operated as a batch burner on a routine basis for the last 18 months and is presently being automated. The automation consists of the furnace temperature control loop and a burner control system. The burner control system is a repeating unit of automatic temperature control (by regulating the fluidizing oxygen flow) and a series of programmed events. The programmed events control fluidizing gas flow, batch product dump valve, batch pneumatic feeder, and refluidizing oxygen flow until the automatic temperature control loop takes over and the sequence starts again. This control cycle is repeated automatically and provides a batch-continuous operation. Although the endothermic burner is similar to the exothermic burner, the fines are burned by containing them within the burner. The low fluidizing velocities used during the endothermic burn stage allow burning the bed to less than 1 percent carbon.

Feed

The feed to the endothermic burner is the product from the exothermic burner. The size distribution of this feed is highly variable, depending on the flowsheet being processed, but at all times it is easily fluidized. The average particle size of the feed varies from 200 to 400 μm for the various flowsheet varieties.

Heat transfer

The burn rates achieved in the 4-in. endothermic burner are about one-half that of the 4-in. exothermic burner, because the bulk of the heat transfer occurs in the transport disengaging height. Also, the operating period in the endothermic stage of burning is a slow burning process. Burn rates of 25 to 35 g carbon/hr-ft^2 are achieved during the exothermic burn period and 5 to 10 g carbon/hr-ft^2 in the endothermic burn period. An average burn rate of about 20 to 25 g carbon/hr-ft^2 or 20 to 25 g carbon/min is achieved during batch-continuous operation.

REFERENCES

1. Nicholson, E. L., et al. Burn-Leach Processes for Graphite-Base Reactor Fuels Containing Carbon-Coated or Oxide Particles. Oak Ridge National Laboratory. U. S. Atomic Energy Commission. Oak Ridge, Tenn. Report Number ORNL-TM-1096. 1965.

2. Steward, H. B., et al. Utilization of the Thorium Cycle in the HTGR. In: Proceedings, 4th Geneva Conference on Peaceful Uses of Atomic Energy. 1971.

3. Blanco, R. E., L. M. Ferris, and D. E. Ferguson. Aqueous Processing of Thorium Fuels. Oak Ridge National Laboratory. U. S. Atomic Energy Commission. Oak Ridge, Tenn. Report Number ORNL-3219. 1962.

4. Blanco, R. E., L. M. Ferris, C. D. Watson, and R. H. Rainey. Aqueous Processing of Thorium Fuels - Part II. Oak Ridge National Laboratory. U. S. Atomic Energy Commission. Oak Ridge, Tenn. Report Number ORNL-3418. 1963.

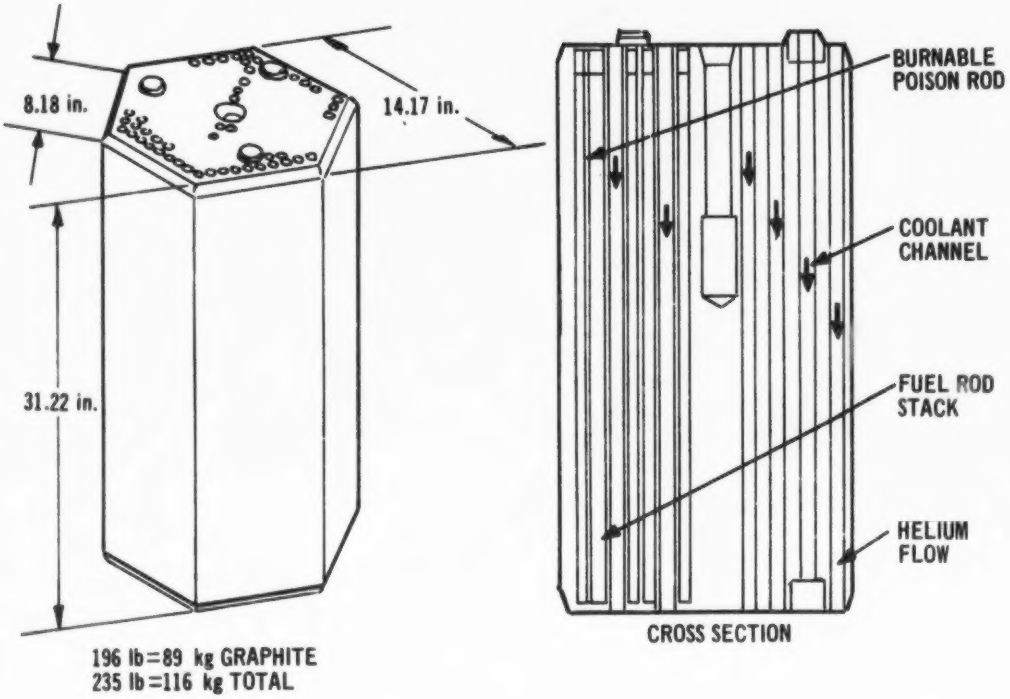


Figure 1. HTGR fuel element.

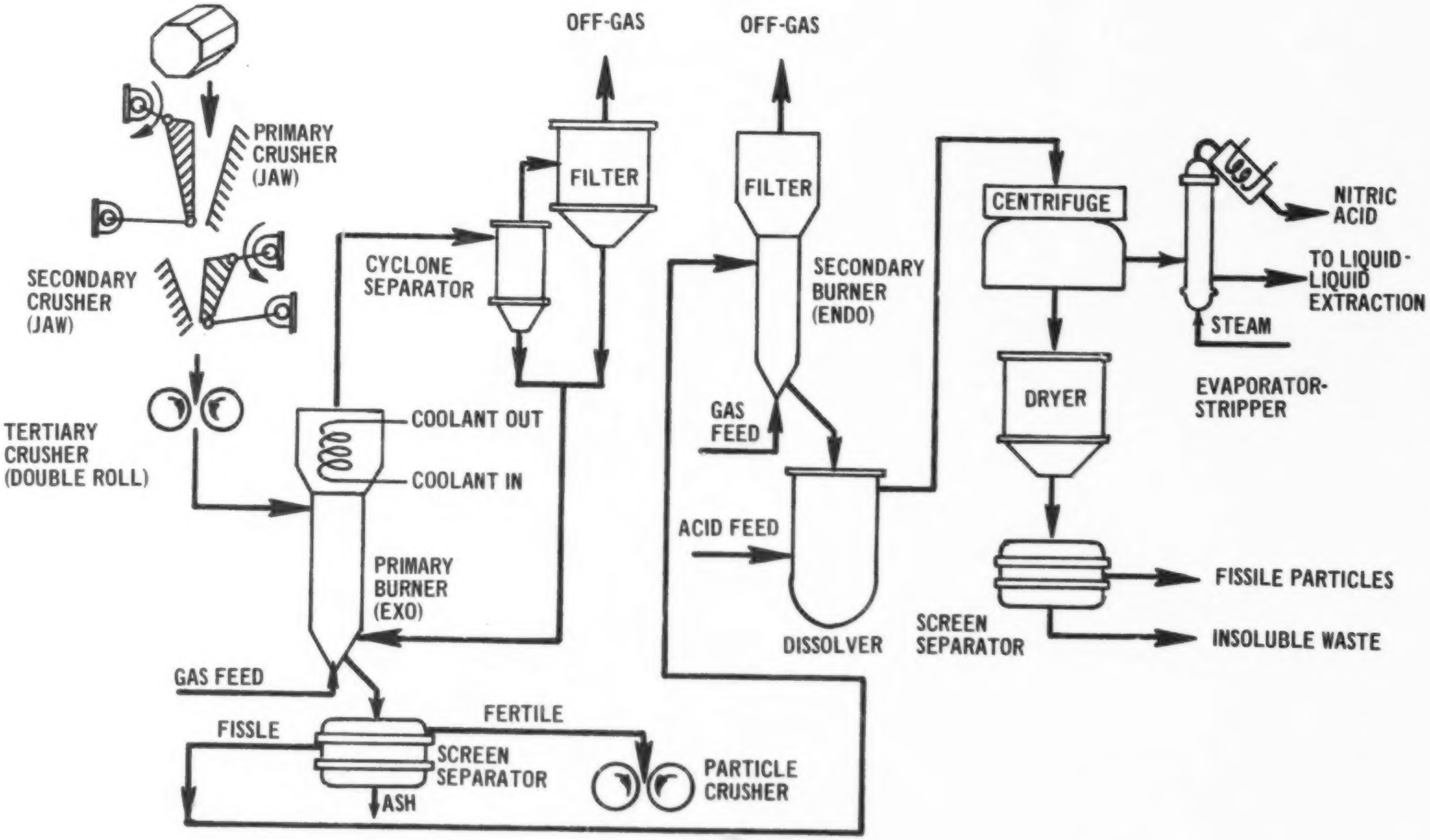


Figure 2. Head-end reprocessing simplified flow diagram.

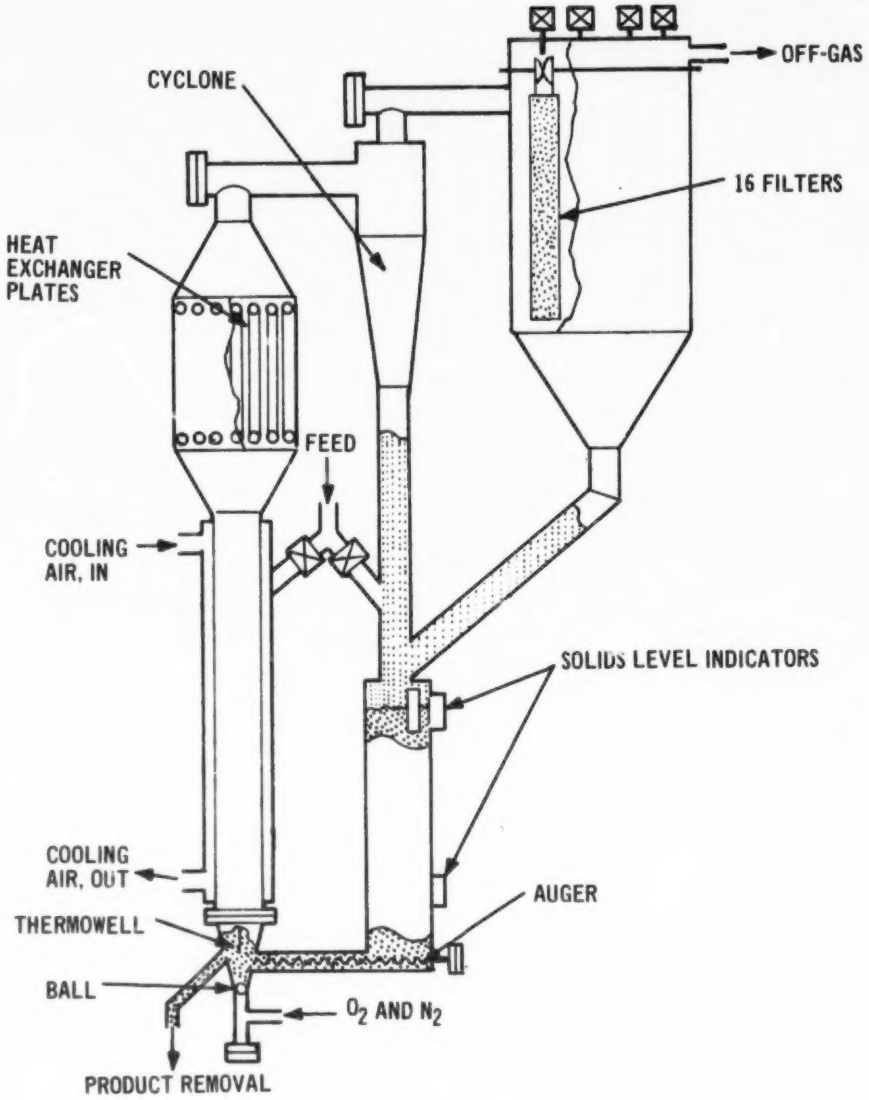


Figure 3. Exothermic fluidized-bed burner.

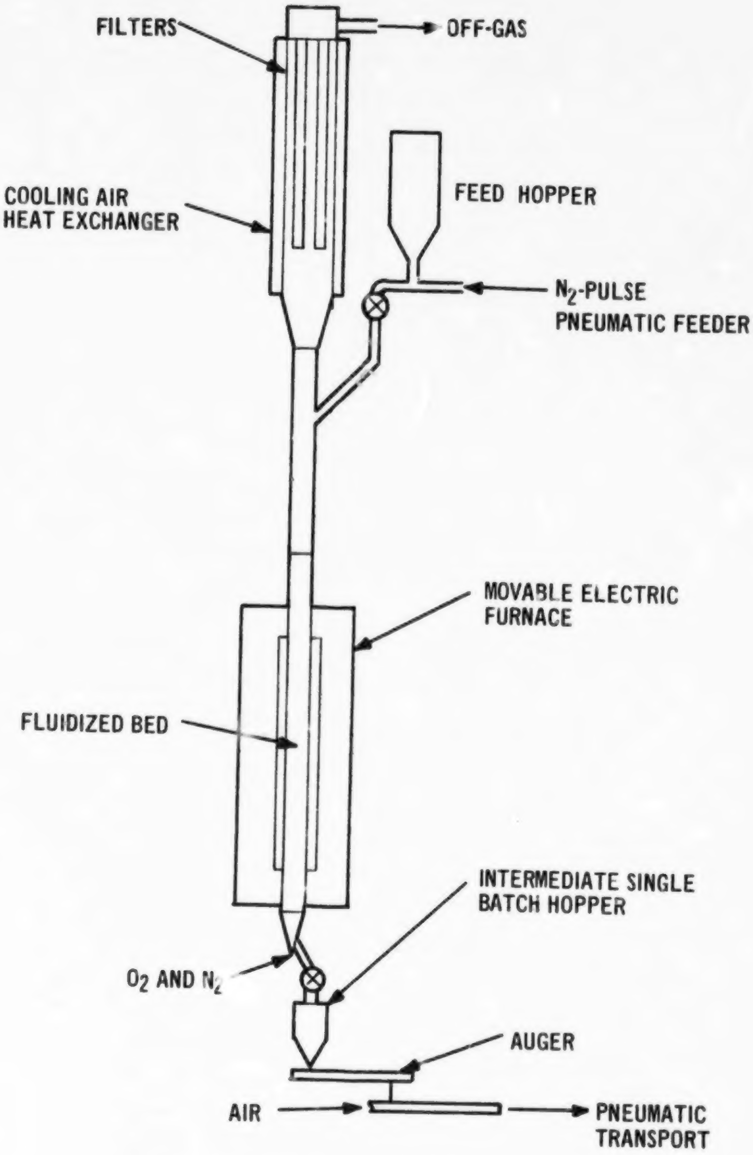


Figure 4. Endothermic fluid-bed burner.

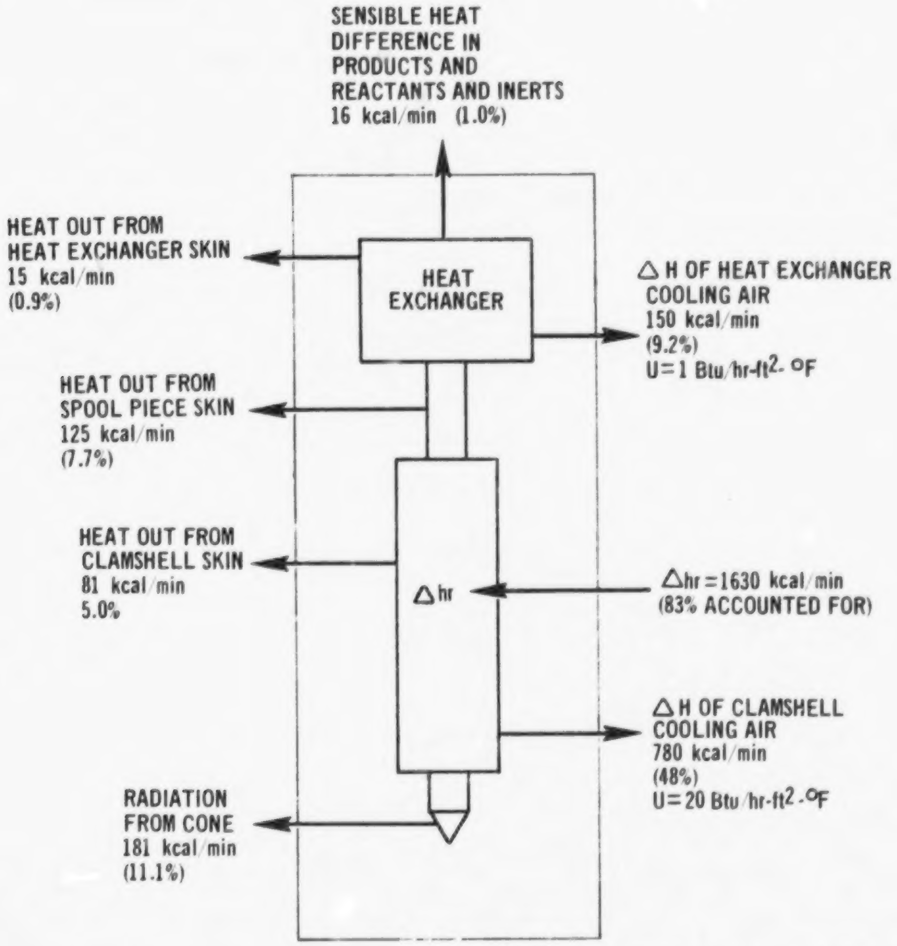
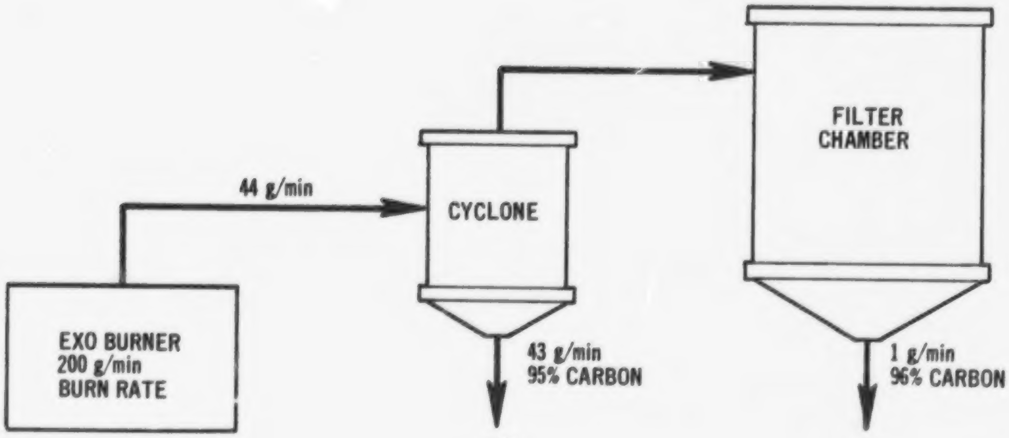


Figure 5. Heat balance and heat transfer coefficients for a typical 8-in. exothermic burner run.



	wt%	wt%
-250 μ+125 μ	0.25	0.0
-125 μ+74 μ	0.25	0.0
-74 μ+44 μ	4.4	4.0
-44 μ	95.1	96.0

Figure 6. Elutriated fines description.

5. STUDIES ON THE COMBUSTION OF NATURAL GAS IN A FLUID BED

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ABSTRACT

Natural gas has been burned with air in a fluid bed of 1.4 ft^2 cross-sectional area, using expanded alumina of 14 to 16 ASTM mesh. The gas/air mixture is supplied premixed. Initial problems of operation, now solved, concerned rapid ignition, and uniform distribution. Light-off initially required two hours or more before combustion was uniform throughout the fluid bed. This period is now consistently 5 to 10 minutes. With very uniform gas distribution, obtained with a distributor of novel design, fluid bed depths were reduced from 6 to 1 inch for complete combustion, even up to superficial (hot) flow velocities of 12 ft/sec. Combustion intensities at 2000°F and 100 percent excess air were in the region of 10^6 Btu/ft^3-hr based upon bed volume. Experiments were carried out with excess air ranging from 60 to about 150 percent, with extinction at the higher value. Gas rates ranged from 6 to 11 ft^3/min. Bed temperatures ranged from 1700 to 2400°F, rising with fuel/air ratio. Air rich extinction boundaries were mapped over a range of fuel/air ratios. Bed temperatures, gas analysis, and pressure drop on a vertical axis through the bed have also been measured. Superadiabatic temperatures and a lowering of the lean flammability limit have been observed. These two observations are explained qualitatively by the preheat effect of the fluid-bed particles. Reaction rates are significantly faster than for free-burning gas in a premixed flame at comparable temperatures and gas concentrations. The effect is attributed to the bed particles. Data on the physical behavior of the bed, with good theoretical agreements, are also given.

INTRODUCTION

This paper describes experiments on the combustion of natural gas in a fluid bed to investigate problems of ignition, even distribution, and combustion speed. Combustion applications of fluid-bed technology can include incineration, steam raising, possibly heating of crucibles, billets, etc. However, use of gas in such applications can present problems.

Fluid-bed combustion of natural gas is, according to conventional belief, beset with difficulties by comparison with combustion of oil or coal. Reasons generally cited are: (1) excessive initial heat up time of the bed, up to 24 hours in the case of very large units; (2) inefficient (incomplete) combustion in the bed, leading to (3) excessive freeboard (overbed) temperatures from final burnup; (4) high bed temperatures, said to be necessary compared with liquid or solid fuels to allow an adequate margin of safety above the commonly accepted ignition temperatures for gas; resulting in (5) excessive thermal stress on heat exchangers included to improve the overall thermal efficiency.

The most significant problem is combustion efficiency, once light-up has been completed. In large units it is customary to avoid premixing the gas and air, because of the explosion hazard; so the gas is supplied directly to the bed through one or more supply ports. A considerable fraction of the gas may then bubble to the surface (2) and, in burning overhed, generate the excessive freeboard temperatures (3) that can overstress heat exchangers (5). Such bubbling behavior parallels remarkably the behavior of coal volatiles, if a high volatile coal is fed too quickly into a fluid bed at too restricted a point.

As explanation of the incomplete combustion inside the bed, the bubbling effect suggests immediately that it would be due to inadequate mixing before the gas breaks the surface. This view is substantiated by estimates of transit time through the bed which considerably exceed the expected reaction times. Nowhere, however, were we able to find any direct substantiation of this expectation in any published literature on gas combustion in fluid beds. Indeed, information on this topic is conspicuous by its absence. There were a few references available indicating that gas could be burned in fluid beds, but not very satisfactorily; and nowhere was there any source of quantitative data, so far as we could establish, that could serve as any basis for engineering design involving commercial use of gas in fluid beds. Furthermore, although our findings as reported here have now substantiated the expectation of very fast reaction in the bed once the gas and air have mixed, at the outset of the project there were some few indications that slow mixing might not be totally responsible for the incomplete combustion in the bed. The argument was based on the usual behavior in the bed immediately after light-off. The bed acted as a flameholder with all reaction above it and no reaction whatever inside it until the whole bed had heated up to a definite temperature. Clearly, the cold particles were providing thermal quenching and/or chain termination. Equally clearly, once hot, the particles could be expected to reverse their action and become sources for thermal and/or chain initiation of reaction. However, this still did not rule out the possibility that the presence of very large surface areas, even of hot solids, in the middle of the reaction zone might so significantly alter the reaction mechanism that the reaction rate could still be appreciably hindered or accelerated. Our findings do, in fact, suggest that the reaction rate may be increased; there are also indications of some other interesting aspects of behavior, such as wider combustion limits and super adiabatic temperatures.

Specifically, however, our starting point was the problem of even distribution and combustion. In the process of investigating this, in conjunction with developing an alternative to the conventional distributor plate, we found a means of reducing the light-off time. The unit thus developed, having very even gas distribution and good fluidization, was then suitable for more detailed measurements of gas temperatures and analyses in the bed, leading to the results indicated above.

What we have to report therefore are potentially valuable data for engineering design and data of more fundamental significance but, withall, not without relevance to design. This is, we believe, the first public report on quantitative behavior of gas burning in a fluid bed.

PRELIMINARY EXPERIMENTS

The first experiments carried out, summarized here, led to a new design concept described below. The unit used for these initial experiments utilized a square, 3/8-in. steel distribution plate, of 13-in. side, carrying 81 perforated studs (a 9x9 array on 1.5-in. centers). The walls were uncooled, constructed of 2.5-in. series superduty (Rockspar) fire brick. Air and gas were supplied separately to mix in the bed. Air was supplied via an air box under the distribution plate, flowing into the bed through 64 of the distributor studs. Gas was supplied through the remaining 17 studs which were connected by a progressively bifurcating line from a 15 psi supply point. The perforations in the distributor studs were 1/8-in. diameter, drilled horizontally from the outside to meet hollow centers. The bed material mostly used was Type 8-F blown alumina of 6 to 12-in. depth in the initial experiments. Instrumentation included: gas and air meters, wall thermocouples, wall pressure taps, suction pyrometer for gas temperatures, and sheathed thermocouples for bed temperatures.

With the unit as described, the bed fluidized satisfactorily in cold flow; but light-off, with combustion predominately in the bed, was never initially achieved. The gas all burned above the bed with the bed acting as a flameholder. It was clear that mixing was the problem because the flames stabilizing on the top of the bed appeared in a set of rings of flame whose pattern was determined by the layout of the gas supply studs. Various means of overcoming this were tried. Unsuccessful or partially successful means included: other bed materials including a fine sand that cascaded through the stud perforations into the wind box; and rotary stirring of the bed by mechanical and pneumatic means. (For the latter, a set of four supplementary air pipes were lowered into the bed to produce rotation of the bed by horizontal jets aimed at an imaginary circle. This was partly successful.)

Success was finally achieved by resting the fluid bed on an underlying bed of limestone rocks (Type 2B) 4.5 inches deep. This was found to be an excellent mixer and distributor, and with this arrangement light-off became possible with reaction ultimately drawn back into the bed. (It was at this point that most of the instrumentation was added.)

Initially, however, light-off was still the excessively long process generally claimed, taking anything up to 2 hours (for a 6 to 12-in. bed). Furthermore, as the flame was drawn into the bed it exploded in a random sequence of strikebacks followed by blow-off (i.e., with the flame oscillating between the top and bottom of the bed). In some instances, the explosions were violent enough to displace some of the wall bricks even though they were mortared into position and partly held there by steel angle frames. Once hot, the bed burned evenly without explosion.

Light-off, however, was ultimately reduced to 10 minutes or less (3 to 4 minutes is about the shortest time so far). It was clear that the flame would not strike back into the bed until the whole of the bed had reached a temperature that would permit it. (The exact temperature is not known.) The heatup took a long time because the particles were only heated at the top of the bed (where the bed top acted as a flameholder) and were cooled again by the cold fluidizing air and gas as they mixed back into the bed. The solution was to fluidize the bed in progressive stages and, with correct gas and air settings, the process is virtually automatic. The gas and air in stoichiometric proportion are initially set at about 10 percent of the rate required for incipient fludization in the cold; the gas is lit to burn over the bed with the bed acting as a flameholder. The gas and air rates are then promptly increased to about half of the cold fludization requirements. The over-bed flame heats the top (unfluidized) layers of the bed and the rising gas and air. When the top bed particles are hot enough, reaction can then start just inside the top of the bed. The consequent local rise in temperature of the gases increases their velocity to above fluidization velocity, fluidizing the top layer of particles. The same process then operates to fluidize the next static layer; this is then repeated, with smooth, non-explosive progression of the fluidized interface down to the bottom of the bed. The gas and air supplies are then adjusted to the required operating levels, and start-up is complete. There is no reason why the same procedure could not be used to cut light-off time on a commercial-scale bed by an order of magnitude, or more.

Solution of the even distribution and rapid light-off problems were the main tasks undertaken in these preliminary experiments. Work was then started on combustion behavior in the bed itself, with initial indications that reaction was substantially slower than it would be in free-burning gas at the same temperature. However, problems due to leakage, mainly through patched cracks produced during the explosive ignition tests, made measurements erratic. An entirely new bed was therefore constructed for further measurements, as described in the equipment section.

Preliminary to the reconstruction, however, some cold model tests of air distribution and fluidization were carried out to aid design.

(1) The first model used water as the fluidizing medium, fed into the bottom of a 12-in. high, 3.5-in. diameter, Plexiglas cylinder, through a 1-in. pipe containing five 4-in. sections of 3/8-in. copper tubing to serve as flow straighteners. An air line to a hyperdermic syringe in the center of the flow straighteners served as a fine bubbler for visual flow tracing in the mixing studies. The fluid bed was simulated by glass beads on a wire screen, about 1 inch above the 1-in. inlet pipe. Beads of 3, 4, and 5mm diameter were used. With water supply up to 6 gal./min., the behavior of fixed, fluidized, and spouting beds could be observed.

(2) The second unit was an air model of a 3-in. diameter feed pipe feeding into a square clay pipe, of 13-in. side, or into 6-in. diameter Plexiglas pipe, again using glass beads to represent the distributor with finer beads or actual bed material above.

These studies indicated the importance of uniformity of the underlying distributor rock, but provided the information to indicate that a single supply point of relatively narrow diameter compared with the fluid bed diameter could be used as long as the distributor rocks (or coarse particles) were deep enough.

EQUIPMENT

The present unit is illustrated in Figure 1. It consists essentially of a refractory lined cylinder of castable refractory, cast inside two oil drums, standing on a welded steel platform 30-in. high. This construction provides an outside diameter of 22 in. and a height of 47 in. The shell is wrapped with 540 feet of 3/8-in. copper tubing for cooling and monitoring the wall loss. Walls, floor, and roof are 3-in. thick, of Hydrecon Tabcast, a 3600°F erosion-resistant castable refractory. The air is supplied to the unit from two staged 24-oz. blowers (360 scfm capacity) through a 3-in. diameter pipe cast in the floor, and the exhaust gases leave through a metal cased flue liner, 12-in. diameter and 2 ft long, leading into a 16-in. diameter stack.

The bed is blown alumina, of mesh size -14 + 16 ASTM, resting on a 6-in. deep bed of crushed refractory, of mesh size -3 + 5 ASTM. At the top of the 3-in.diameter air supply pipe, the crushed refractory is supported on a 1/16-in. perforated steel plate. The crushed refractory provides all necessary air and gas distribution across the full width of the unit.

An ignition burner, at a height of 22 inches, is provided for startup and safety. To observe the bed, two inspection ports closed with 2-in. Vycor discs are provided in the top; the top also contains two holes for insertion of probes. For access to the bed, the top can be removed using a 1/2-ton differential chain hoist.

The air and gas are fed to the bed through a mixer, after being metered by rotameters reading to 280 and 15 scfm full scale, respectively. The mixer is a 2-1/2-in. diameter (Pyronics) venturi unit, followed by a Tee to carry the mixture into the bottom of the bed; the other leg of the Tee provides a sump for any material falling through the inlet hole. To permit increased flow rate through the bed, if required, a bypass valve is provided around the air rotameter, with an "Annubar" flow meter to measure these higher flow rates.

Cast into the furnace at heights of 9 and 12 inches above the base are two rows of 11 holes capped with 1/4-in. pipe nipples for instrument access or solid feed. Twenty-three pressure tap holes lined with 1/4-in. porcelain are cast into the side at 1/2-in. intervals from the bottom to a height of 8 inches, and every 1 inch thereafter to a height of 16 inches. Before casting, 1.5-in. pieces of stainless steel were soldered to the oil drum shells at all pressure tap and thermocouple stations to protect the porcelain from breakage. Shielded thermocouples flush with the inside wall are provided at elevations of 1, 3, and every inch thereafter up to 19 inches. Two sets of thermocouples are also mounted at intervals of 1/2-in. depths into the wall to monitor the wall temperature profile. All thermocouples are Chromel-Alumel, with read out by a Leeds & Northrup 24-point recorder. Pressure is monitored by a 36-in. tube, well type manometer. A Chromel-Alumel thermocouple is used for bed temperature measurements with readout by a single channel Honeywell recorder; a check on bed temperature can also be made by optical pyrometer.

A draft gauge with range from +0.005 to -0.015-in. wc is connected to the horizontal flue-run for pressure monitoring. Two water cooled probes 48-in. long are used for gas sampling, one in the stack and the other in the bed. Gas is continuously monitored for CO_2 and CO with infrared instruments, and for O_2 with Thermox analyser using a fuel cell as the sensor element. Incomplete combustion of the gas is also determinable directly by an MSA total combustibles analyser.

Initial startup, when the unit was first completed, was accomplished without difficulty using the technique described earlier. However, some difficulty was initially experienced in maintaining fluidization for any length of time. The bed would start to blind at some point. Fluidization would be lost; the gas/air flow would be diverted from those regions which would cool, thus tending to prevent refluidization. Careful analysis of the problem suggested that it was probably due to fines in the bed material. At all events, removal of fines by screening on a 16-ASTM sieve eliminated the problem.

Progressive improvements to the design and method of operating the unit resulted in a steady improvement in fluidization; as fluidization improved, the bed depth required for complete combustion dropped steadily. In the preliminary experiments on the first unit, bed depths up to 12-in. were used. In the current unit, beds were initially 4 to 6-in., but were finally reduced to 2 inches and then to 1 inch with the even fluidization achieved. For heating purposes (in the bed) or solid wastes incineration greater depths are required. For our immediate purposes here, however, since no interesting combustion behavior occurred above the bottom 1 or 2 inches of the bed, the top layers were "omitted" to allow easier access by probes to the regions of interest. It should be emphasized, of course, that until the present unit was completed, and the very uniform fluidization obtained, the evidence indicated that combustion required 3 to 6 inches.

PHYSICAL PROPERTIES OF THE FLUID BED

In the course of the combustion investigations, the porosity and other properties of the hot bed were measured, some of which were obtained as a matter of necessity. Because of their potential engineering value for design they are summarized here. It is emphasized that these are measurements made at high temperatures, up to 2400°F, with combustion following; since the bed depths were small, the data should be valid for all bubble free conditions. The data of principal interest were the expanded bed heights, porosities, gas velocities, and residence times, all of which are needed for interpretation of the combustion data.

Values of pressure drop (ΔP) between the top of the bed and a point below the top of the bed were first measured by traversing the bed with an open-ended, water-cooled probe. (The kinetic head contribution is too small to cause any determinable error.) Measurements were taken for six different fluidizing velocities, with bed temperatures ranging from 1800 to 2350°F; the normalized results of ΔP against h are shown in Figure 2 where h is measured from the bottom of the bed. The normalizing parameters used were the total pressure drop ΔP_t and the (expanded) bed depth L. The top of the bed was identified by a break in the slope of the ΔP against h line. At first glance it is evident that the plot of Figure 2 is respectably linear, in agreement with theoretical predictions quoted in standard tests; e.g., 1, 8. With closer inspection, the slight curvature of the line is self-evident; the curve is probably due to the temperature variation and hence the velocity variation through the bed. However, the departure from linearity is small enough that it can be neglected for our purposes.

The normalizing parameters, ΔP_t and L_0, were also found to obey simple theory. Equations given by Davidson and Harrison[1] have been used. Renormalizing their equation (1.11) against the initial bed depth (L_0) at incipient fluidization velocity (U_0) when the initial porosity is ε_0, we obtain

$$\left(\frac{\Delta P_t}{\Delta P_{ot}}\right)\left(\frac{L_0}{L}\right) = \left(\frac{U}{U_0}\right)\left(\frac{\varepsilon_0{}^3}{\varepsilon}\right)\left(\frac{1-\varepsilon^2}{1-\varepsilon_0}\right) \quad (1)$$

where ΔP_{ot} is the pressure drop across the bed at incipient fluidization. However, $(\Delta P_t/\Delta P_{ot})$ is unity in the fluidization region, as Figure 3 illustrates. Note also that the value of ΔP_{ot} is below that required to balance the bed weight. This is in accordance with Trevedi and Rice's experiments.[5] A further simplification of equation (1) is possible by using equation (1.22) of reference 1, again renormalized to yield

$$(U/U_0) = (\varepsilon/\varepsilon_0)^3(1-\varepsilon_0)/(1-\varepsilon) \quad (2)$$

Figure 4 substantiates this equation, illustrating the linearity obtained by plotting $\varepsilon^3/(1-\varepsilon)$ against U, where U is determined under the hot condition. Figure 4 also shows the actual variation of ε with U. Values of ε were calculated from the particle denisty (σ) and the bulk bed density (p) (calculated from bed weight, depth, and areas) using

$$\varepsilon = 1 - p/\sigma = 1 - 0.464\, L_0/L \quad (3)$$

The incipient fluidization porosity (ε_0) is 0.536, which is somewhat above the value of 0.476 for cubic packing of uniform spheres. Since the alumina particles are quite good spheres it is not unrealistic to assume this less dense packing is due to particles bridging void areas.

Substituting equation (2) in equation (1) yields

$$L/L_0 = (1-\varepsilon_0)/(1-\varepsilon). \quad (4)$$

This relation is substantiated by Figure 5. The slope, equal to $(1 - \varepsilon_0)$, has a value 0.46 yielding $\varepsilon_0 = 0.54$, which is good agreement. Figure 5 also shows the variation of (L/L_0) with U, with the fitted curve calculated from equation (2) adopting the experimental values of U_0 and ε_0. The gentle curve could be well approximated by a straight line, which is a consequence of $\varepsilon^3/(1-\varepsilon)$ varying almost linearly with $1/(1-\varepsilon)$; it is approximately proportional to U.

Absolute calculation of the incipient fluidization velocity is not quite so satisfactory although the experimental value can be bracketed. Again, using an expression given by Davidson and Harrison,[1]

$$U_0 = 0.00081\ (\sigma g d^2/\mu)\ (\text{cm/sec}) \qquad (5)$$

where d is the particle diameter (=0.0510 in.) and μ is the dynamic viscosity. U_0 was calculated for ambient temperature and 1800°F, knowing the average weight of one particle. The predicted values were 3.28 ft/sec and 1.21 ft/sec (at the higher temperature). This bracketed the experimental value of 2.0 ft/sec. Since the numerical factor of 0.00081 was determined for experiments on fluidizing with water,[1] the agreement is acceptable.

From the data given, calculation of the actual (as opposed to the superficial) velocity is straight forward using a porosity correction; the same occurs for the residence time in the bed (τ_s). The results of the two calculations are given in Figures 6 and 7, respectively, with bounding values for ϵ_0 and $\epsilon = 1$ included for comparison. The residence time data, in particular, are needed for discussion of the combustion behavior.

The general agreement with theory established here would support the use of the equations tested for engineering design and scale-up, if used with care.

COMBUSTION BEHAVIOR

General

The problems of light-off have been fully covered above. Once lit, combustion can be maintained indefinitely as long as no blinding of the bed by fine particles occurs. In the earlier experiments, when fluidization and mixing of the fuel and air were relatively poor, the over-bed gases were periodically flecked with yellow as bubbles of gas broke the surface and burned in the freeboard space. This behavior was progressively eliminated by improved fluidization and mixing. With the present arrangement, utilizing premixing in a venturi mixer, there can be no bubbles of fuel rich gas. However, it was clear that the speed of reaction was still strongly influenced by the fluidization quality. When this was poor, 4 to 6 inches were apparently required for combustion. As fluidization was improved, the space required was progressively reduced until, as mentioned above, it could be accomplished well within a 1-in. bed. Under these conditions, all that can be seen through the sight glass in the top is the red hot bed-top in continuous motion without any spouts, and with particles welling up and disappearing again.

In the experiments next described there were three objectives. With the expectation that gas-fired fluid beds will be increasingly used in commercial practice, attention was first directed at two aspects of safety: (1) if fuel to a bed is cut off, at what minimum temperature will it relight? and (2) if a surge of air or neutral gases leans out the fuel-air mixture, at what gas percentage and bed temperatures will there be extinction? With information on these first two, attention was then given to behavior in the bed in an attempt to determine whether combustion in a particle-filled volume is affected in any way by the presence of the particles.

Relight

To investigate relight behavior, the procedure was to set up the bed in normal operating condition and then to switch off the fuel and the air, or to decrease the air. The bed would cool; and periodically at recorded temperatures the fuel supply would be restarted. The observation then made was whether or not the bed temperatures would start to rise again. This was taken as a condition of relight.

The experiments on relight were carried out at a fairly early stage in the investigation, with beds 4 to 6-in. deep, and mostly at fuel-air ratios closer to stoichiometric than to the lean limit. Under these conditions, relight was always successful down to bed temperatures of 750°F (400°C). Lower temperatures than this were not investigated for safety reasons. This is substantially below the values generally listed in standard data tables; e.g., 4, for auto (spontaneous) ignition temperatures. Reference 4, for example, quotes: 1290°F (700°C) for the stoichiometric mixture; greater than 1200°F (650°C) for the most easily ignited mixture; and adds the comment that under pressure, the temperature is never less than 880°F (470°C). It seems fairly clear that the method of determination is too different from a fluid bed for the results to be relevant.

Extinction

The procedure for determining extinction, at the low limit, was to start with the bed in normal operating condition at some suitable fuel and air rate, and then to lean out the mixture by stepwise reduction of the fuel rate. When the flame extinguishes the temperature falls rapidly. The extinction point can only be judged between two steps, always approached from the flame side of the boundary. After extinction the bed was relit, the air rate reset, and the sequence repeated.

A typical set of results is shown in Figure 8 which illustrates plots of temperature (maximum observed by thermocouple) against the superficial velocity (calculated utilizing the maximum temperature) through the bed for several run sequences. The extinction-temperature boundary is clearly marked as a heavy dashed line. The lightly dashed lines represent constant fuel-air ratio in percentages by volume. The continuous line marked 5.3 percent is the conventional low limit. It can be seen that a considerable number of combustion points lie below the low limit. This is more clearly seen on Figure 9, showing continued combustion down to 4 percent methane, more than 1 percent below the normal low limit. Figure 9 also includes the theoretical adiabatic flame temperature line with some temperatures exceeding the adiabatic value. The source of these unexpected peculiarities lies in the heat exchanger effect of the fluid-bed particles, as discussed below.

Bed Profiles

The same super-adiabatic behavior also occurs in the bed itself. Figure 10 is a typical temperature traverse down through the bed. Traverses were made both with sheathed and bare thermocouples, and a displacement of the profiles was observed. By using a special sheathed couple set at right angles to the holder, it was established that conduction down the sheath could result in spuriously high temperatures at a given point. The data are, in effect, translated by about 1/4-in. However, this may not entirely account for the differences. Temperatures exceeding the theoretical adiabatic by 50 to 150°F have been recorded in many of the temperature traverses made.

Clearly, the rate of heat removal must exceed the rate of reaction at locations above the temperature peak (in the regions marked C and D) in order to allow the temperature to decline. Furthermore, the reaction is probably totally completed at the location of the temperature peak. To check this, gas analyses were taken throughout the bed; Figure 11 illustrates one method of representing the fuel consumption calculated from the gas analyses. The graph represents the unburned fuel, on a log-linear plot, declining with distance up through the bed. The fuel unburned was back calculated from the CO_2 analysis; the CO was never found to exceed 0.75 percent and was disregarded in the calculation. The two curves represent two different bed temperatures, 1850°F (1010°C) and 2350°F (1290°C), with fuel burn-up easily followed through the bed, although the times represented are only of the order of milliseconds. The fuel concentration in each case decays more or less exponentially through the bed. Reaction is fast with 90 percent reaction in 16.0 msec and 2.2 msec for the lower and higher temperature beds, respectively. These figures are in agreement with the temperature profiles and with the prediction that combustion is mostly complete before the temperature peak is reached, and all (detectable) combustion is completed within the bed.

DISCUSSION

The physical behavior of the bed is in good agreement with expectation from established theory and requires no further comment. The combustion behavior, on the other hand, shows a number of unexpected features.

(1) The lean limit extension and super-adiabatic temperatures indicated in Figures 8, 9, and 10 were particularly unexpected. They are, however, simple to explain. They depend on a heat recovery or heat exchanger effect due to the bed particles moving up and downstream. To understand the behavior, consider first the effect of a heat exchanger in the exhaust of an otherwise adiabatic flame system. With the heat exchanger not connected, the gas exit temperature of the flame system is the adiabatic flame temperature. If the heat exchanger is allowed to heat the incoming combustion air for the flame system, the gas exit temperature must be boosted to adiabatic plus the preheat. Overall, of course, there is no gain because the extra heat in the exit gases is removed by the heat exchanger (exactly, in a no-loss system); the gases now leave the heat exchanger at the adiabatic flame temperature. This is, of course, no more than the usual application of the heat exchanger although the potential of heat exchangers for producing super-adiabatic temperatures is not always recognized for what it is.

In the case of the fluid bed, the net heat exchanger effect is clearly evident, but it is not particularly efficient in this role since the temperature excess is only 50 to 150°F. The preheat influence is believed to be most marked in the early stages of the temperature rise. In these regions the supply of bed material must be predominantly from above (i.e., from hotter zones), whereas further up in the bed there can be as much material supplied from below as above, thus contributing to cooling of the upper zones. Nevertheless, the overall consequence is to accelerate the rate of heating, and therefore the rate of combustion in the bed.

(2) The same preheat effect is responsible for the extension of combustion below the usual low limit. Again consider the case of air preheated to a very high temperature (say, by a heat exchanger). If the temperature is high enough, any quantity of fuel, however small, injected into that air stream cannot be prevented from reacting completely. Between this extreme condition and the condition of the normal lean limit there is a range of rising temperatures permitting a progressive lowering of the lean limit to zero. Weinberg,[7] for instance, recently quoted a system in which stable combustion is maintained at a gas concentration of methane in air of 1 percent; combustion contributed 250°C and preheat contributed 1000°C.

Clearly, the heat exchanger effect can be at least partly responsible for the widening of the combustion limit found in the fluid bed. However, the magnitude of the effect — a drop of over 1 percent in the low limit — does seem to be rather large for the relatively small temperature increase over adiabatic, of 50 to 150°F. Some other effect, as discussed below, may also be involved. This view is supported by a few measurements using beds of double the depth. The maximum temperature increased by about 50°F, but the limit mixture at extinction was unaffected. It may also be of significance that the super-adiabatic temperatures were only obtained at the higher flow rates, presumably because the heat exchanger effect was stronger.

(3) Indications that factors, other than those already mentioned, could be influencing the reaction were obtained from estimates of reaction time. Table 1 lists some estimates of time to complete 90 percent of the reaction (based on the Figure 11 plots). Included for comparison are some data from other sources.[2,3,6] The most directly comparable

Table 1. ESTIMATES OF REACTION TIME IN A FLUID BED AND IN CONVENTIONAL FLAME SYSTEMS

Investigator	CH_4, %	O_2, %	Inert, %	T_F		V		τ	
				°K	°F	ft/sec	cm/sec	msec	
Present work	4.25	20.1	75.65 (N_2)	1290	1865	2.7	83	22[a]	Total bed residence time
								16[a]	Temperature peak 90% reaction
								11	Total bed residence time using T_{max}
								6	Temperature peak 90% reaction using T_{max}
	5.92	19.88	74.2 (N_2)	(adiabatic) 1650 (observed) 1530	(adiabatic) 2450 (observed) 2300	2.2	74	2.2	Temperature peak 90% reaction using T_{max}
Dixon-Lewis[2]	5.03	19.94	75.03 (N_2)	1528	2300	0.17	5.2	17 ± 3	90% reaction
Levy[3]	5.4	20.0	74.6 (AR)	1950	3050	1	31	4.9 ± 1	90% reaction
Van Tiggelen[6]	Stoichio-metric	Stoichio-metric	73 (AR)	2110	3340	1.1	34	0.060	Mean molecular
Van Tiggelen[6]	Stoichio-metric	Stoichio-metric	65 (AR)	2370	3800	2.2	66	0.015	residence time

[a] Temperature does not show a significant rise until after 8 msec have elapsed.

data are those given by Dixon-Lewis.[2] For 90 percent reaction, 17 ± 3 msec are required in a flat-flame system, while only 2.2±0.5 msec are required in the fluid bed at very close to the same temperature (2300°F), and only 4 ±1 msec for 99 percent reaction. This factor of 6 or 7 difference is clearly significant.

The most probable explanation that comes to mind is, of course, enhanced reaction due to the particles. This could be a result of either initiating more gas-phase reaction or catalytic surface reactions.

(4) Some choice between catalytic surface reactions or enhanced gas-phase reactions may be possible from further data developed from the fuel consumption curves of Figure 11. Assuming that the heat from consumption goes exclusively into the mixture and the products at the local level, a temperature profile through the bed was calculated. Figure 12 shows how this compares with the measured profile. The deviations at the top end have already been explained as a result of the preheat effect. The discrepancy at the lower temperatures, with the predicted temperatures substantially in excess of measurement, was totally unexpected.

In accounting for the observed discrepancies a number of explanations were considered; all but two were discarded. The simplest explanation is that the gas analyses may show spuriously high CO_2 values because of continued reaction in the sampling probe. Against that, however, is the matter of the low temperatures involved so that any substantial cooling of the gases would freeze the composition. The other explanation is more involved but is also considered more likely. It is based on the assumption of significant temperature difference between the particles and gas, which is quite possible considering the rapid translation of hot particles into the cooler bed zones and the short times involved for re-equilibration. If, therefore, the hot particles stimulate surface catalytic reaction so that most of the heat released goes directly into the particles, the lead in the particle temperature above the gas temperature will be maintained until reaction decays. Either bare or sheathed thermocouples will then take up a temperature intermediate between the gas and the particles, but the sheathed couple can be expected to be more responsive to the particle temperature because of the enhanced heat transfer coefficient between particles and a surface. This would then help to explain the discrepancy noted above between the bare and sheathed couples as a factor additional to conduction as noted.

(5) Finally, a brief examination of the relevance of this information to engineering applications is in order. The outstanding point is that reaction in unpremixed systems will clearly be dominated by the mixing behavior; the time for reaction can be virtually ignored unless temperatures are very low indeed. Clearly, future experiments should include measurements down to about 1000°F or lower, which may well be achieved on occasion if a very wet slurry or sludge is incinerated. However, on the unit used, controlled variation of the bed temperature has been very difficult. There could be some advantage in reducing the bed size to provide better bed temperature control. The other aspects of possible engineering significance are the wider combustion limits and enhanced reaction rate (i.e., speed of ignition) due to the hot particles. These could also increase the risk of serious explosion of any large bubbles of premixed fuel and air, if such bubbles are ever permitted to form. The extinction and relight conditions are also important; consequently, development of analytical models for experimental test is now required to provide a more reliable basis for extrapolation. (Two models have been developed, but they are still too limited in their assumptional basis to be of much value yet.) Beyond that, what is mainly needed now for engineering purposes is an understanding of the behavior of jets and mixing in fluid beds.

In conclusion, therefore, the results developed in this paper substantiate the reasonable expectation that reaction of gas in fluid beds is fast, and that problems of incomplete bed reaction must be due to poor mixing in the bed. In addition, and unexpectedly, it was also found that reaction seems to be accelerated by the presence of particles which also can widen the combustion limits and generate super-adiabatic temperatures. Other results include the development of a means of rapid light-off and a demonstration that the physical behavior of the bed is in general accordance with expectation from available theory, in spite of the simultaneous presence of combustion.

ACKNOWLEDGMENTS

We have pleasure in acknowledging support for this work from Consolidated Natural Gas Service Corporation under Grant No. C-70-29-2. We also gratefully acknowledge assistance in the construction of the equipment from Messrs. R. Frank, C. Martin, and D. Simpson. The first author also wishes to thank Mr. M. Kuwata for assistance he so ably provided during this work.

LIST OF SYMBOLS

c_{if} = Initial fuel concentration, by volume

h = Location in bed measured from bottom, in.

L = Bed thickness, in.

ΔP = Pressure drop, in. H_2O

T_F = Flame temperature

U = Superficial velocity (calculated at bed maximum temperature), ft/sec

V = Flame velocity

ϵ = Bed porosity

ρ = Bed bulk density, lb/ft^3

Φ = Completeness of combustion

σ = Particle mass density, lb/ft^3

τ_s = Gas residence time in the bed, sec

Subscript

o = Incipient fluidization values—also used to denote fixed bed values where applicable (ΔP, ϵ, L).

REFERENCES

1. Davidson, J.F. and D. Harrison. Fluidised Particles. Cambridge, Cambridge University Press, 1963.

2. Dixon-Lewis, G. and A. Williams. Some Observations on the Combustion of Methane in Premixed Flames. (Presented at the 11th Symposium (International) on Combustion, 1967, pp 951-958.)

3. Levy, A., J.W. Dredege, J.J. Tighe, and J.F. Foster. The Inhibition of Lean Methane Flames. (Presented at 8th Symposium (International) on Combustion, 1961, pp 524-533.)

4. Spiers, H.M. (ed.). Technical Data on Fuel. Edinburgh, British National Committee World Power Conference, 1962, p. 260.

5. Trivedi, R.C. and W.J. Rice. Effect of Bed Depth, Air Velocity, and Distributor on Pressure Drop in an Air Fluidized Bed. Fluidized Bed Technology, Chemical Engineering Progress Symposium Series, American Institute of Chemical Engineers. 62, 1966.

6. Van Tiggelin, A. and J. Deckers. Chain Branching and Flame Propagation. (Presented at 6th Symposium (International) on Combustion, 1957, pp 61-66.)

7. Weinberg. Combustion Temperature: The Future. Nature, 233:239, 1971.

8. Zabrodsky, S.S. Hydrodynamics and Heat Transfer in Fluidized Beds. Cambridge, MIT Press, 1966.

ADDENDUM ON RELIGHT BEHAVIOR

Since this paper was written further data have been obtained concerning the relight behavior of the bed. This further information amplifies the results quoted in the section titled "Combustion Behavior: Relight."

The method of experiment was as follows. In the tests described in the section on "Relight" the bed was first fired up normally till thermal equilibrium was obtained. Then the gas flow was decreased to a value below the lean flammability limit so that the temperature declined slowly. Combustion was still occurring in the bed, but the heat release rate was insufficient to maintain equilibrium. We also conclude that combustion in the bed was not quite complete, because the reaction was evidently continuing on surfaces in the freeboard area, such as the stainless-steel sheathed temperature probe. It was observed that the probe was glowing red when the bed was black and, therefore, presumably cooler. When the gas flow was increased (to the level of stoichiometric gas-air mixture), relight was always obtained down to 750°F (400°C), as already reported. The glowing thermocouple sheath did not originally appear to be important since the temperatures reported were presumably those of the thermocouple (and sheath); the reported temperatures, being greater than those in the bed, provided a conservative margin.

These tests were recently repeated, but with the gas flow turned completely off. The temperature declined much more rapidly since there was no combustion occurring in the bed to retard the temperature decline rate. This rapid drop made temperature estimation difficult which was the reason for trying to control the temperature rate of fall in the original experiments. There was also no combustion in the over-bed region to provide an over-bed ignition source as in the first case. Relight under these new conditions was not obtained even at 1400°F (750°C).

These results indicate that in the first case the hot areas in the over-bed region were providing the ignition. Thus, for engineering applications, an over-bed ignition source should be present for safety reasons.

The relight temperatures obtained under the second set of conditions are evidently at or above the values generally quoted for auto-ignition. The experiments thus underline the very significant distinction between auto ignition and reactor ignition. They show very clearly the safety of the system and ease of relight down to very low temperatures, even if the gas concentration is very substantially below the low limit. Risk occurs only in the event of total failure of the gas supply, which is easily guarded against by standard safety precautions.

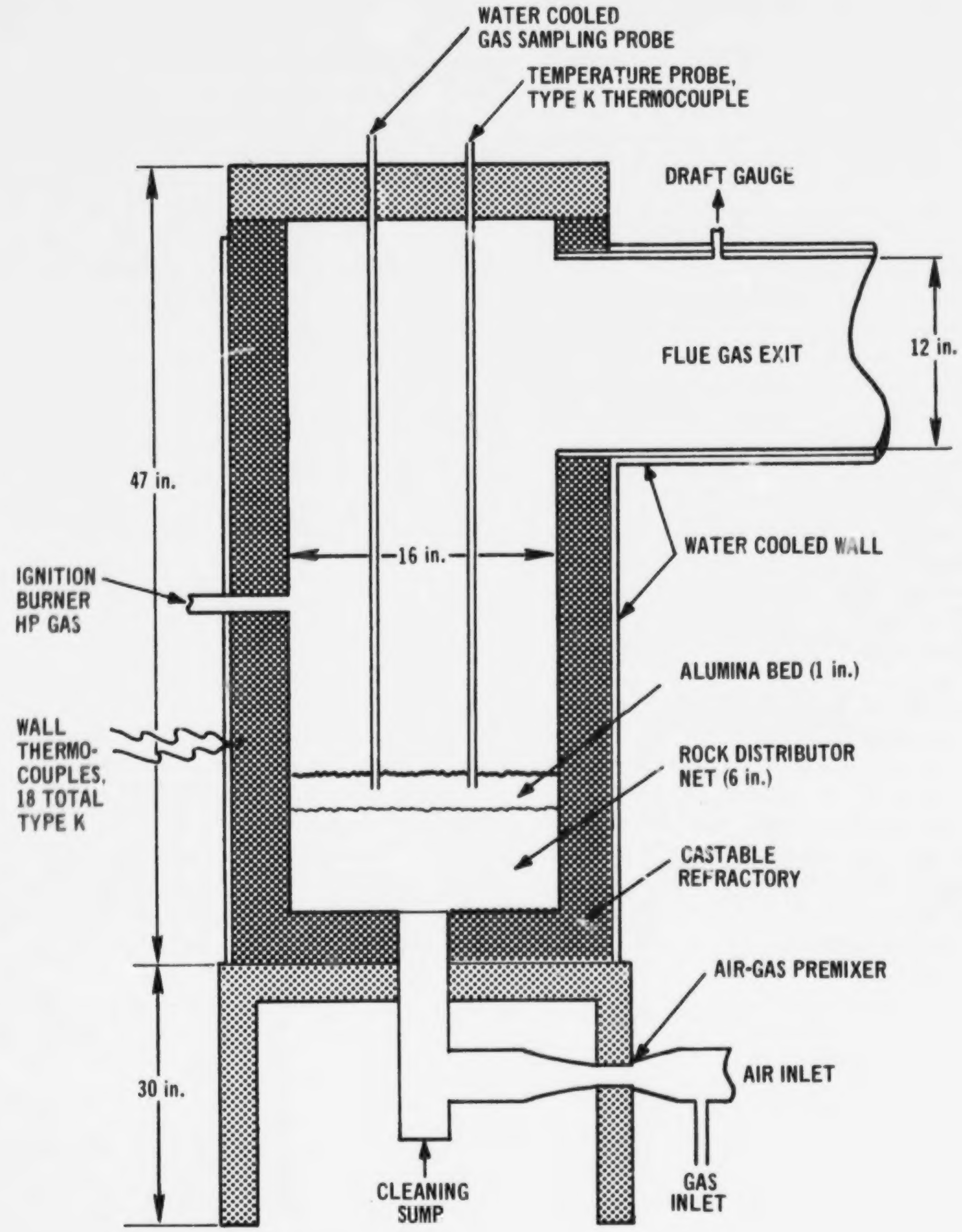

Figure 1. Details of experimental apparatus used for these tests.

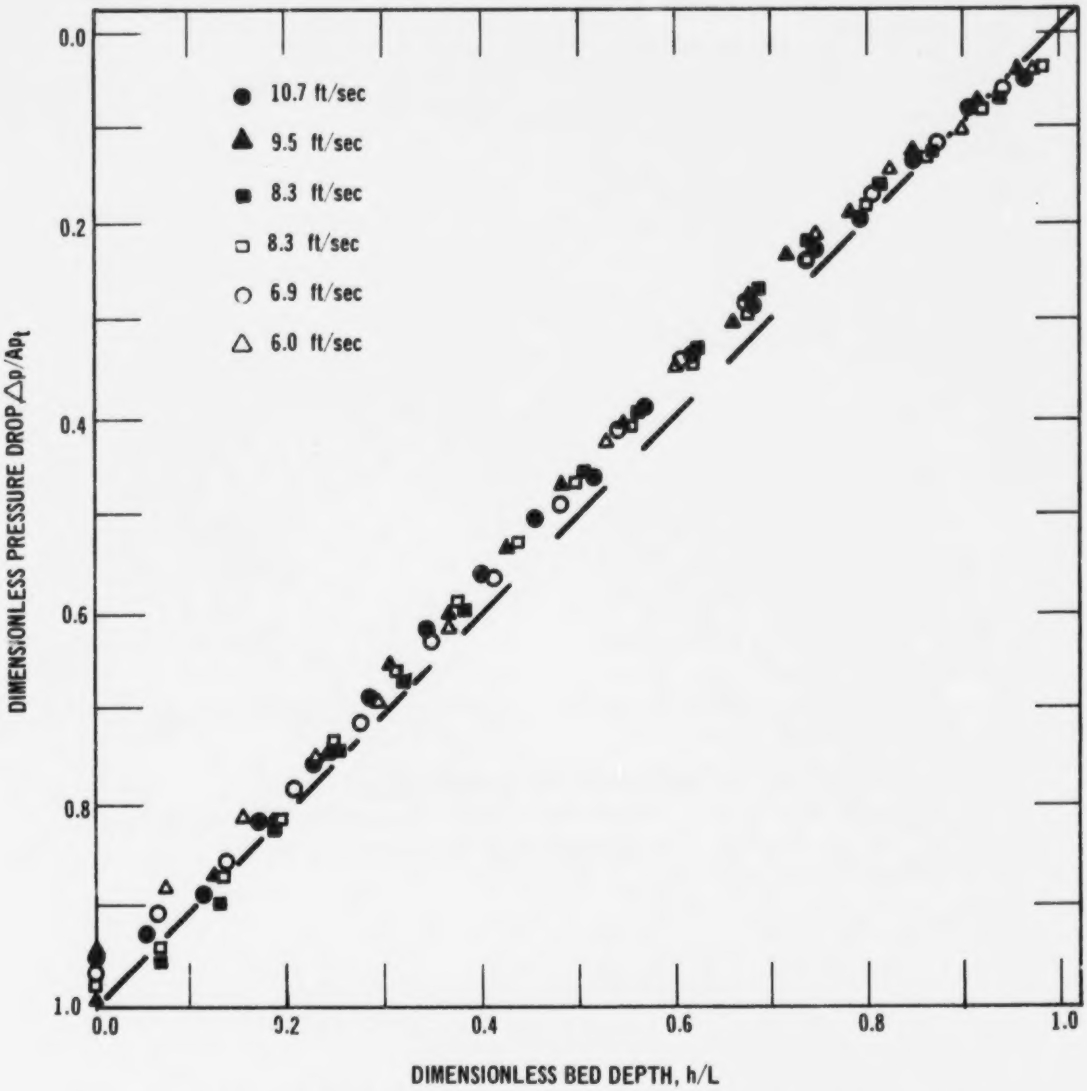

Figure 2. Variation of dimensionless pressure drop $\Delta p/Ap_t$ with bed depth h/L. Data taken with water cooled probe in a thin fluidized bed of 1-in. fixed-bed thickness. Combustion in the bed produced a temperature variation of 1800-2350 °F, and velocities of 3 to 5 times incipient fluidization velocity (2 ft/sec).

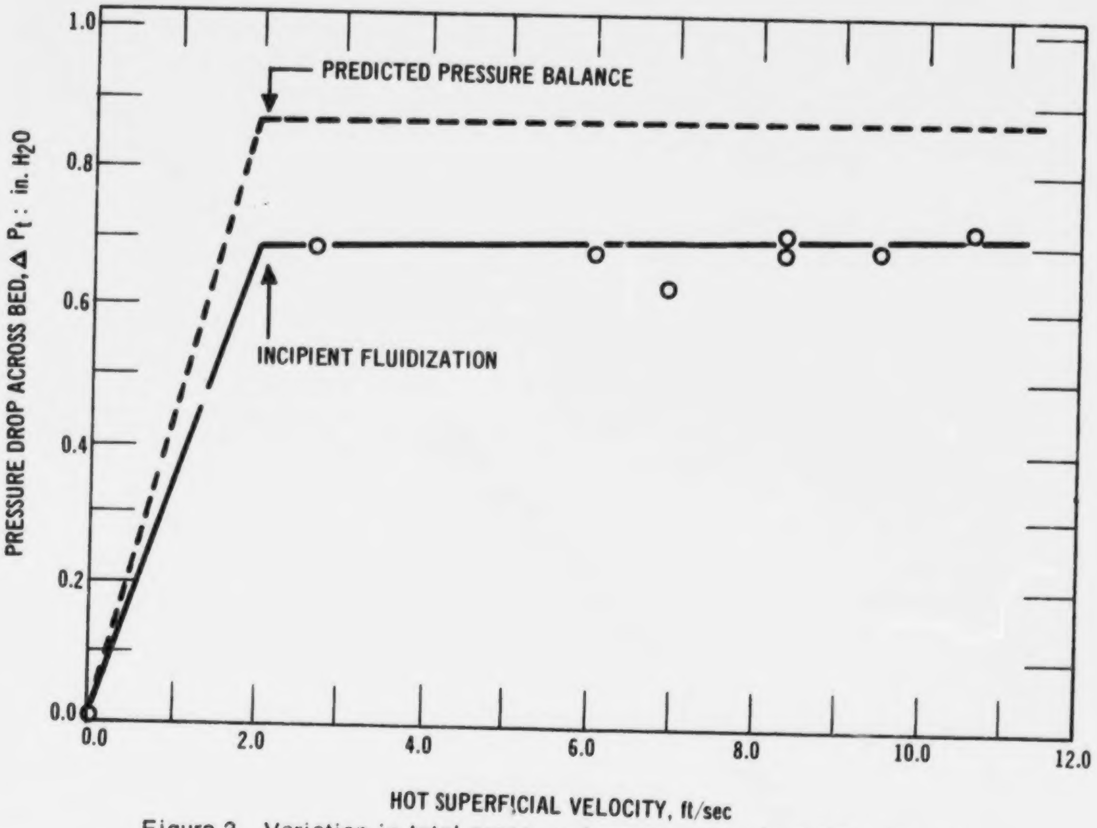

Figure 3. Variation in total pressure drop across the bed ΔP_t with hot superficial velocity. The pressure drop necessary to support the bed weight is also indicated.

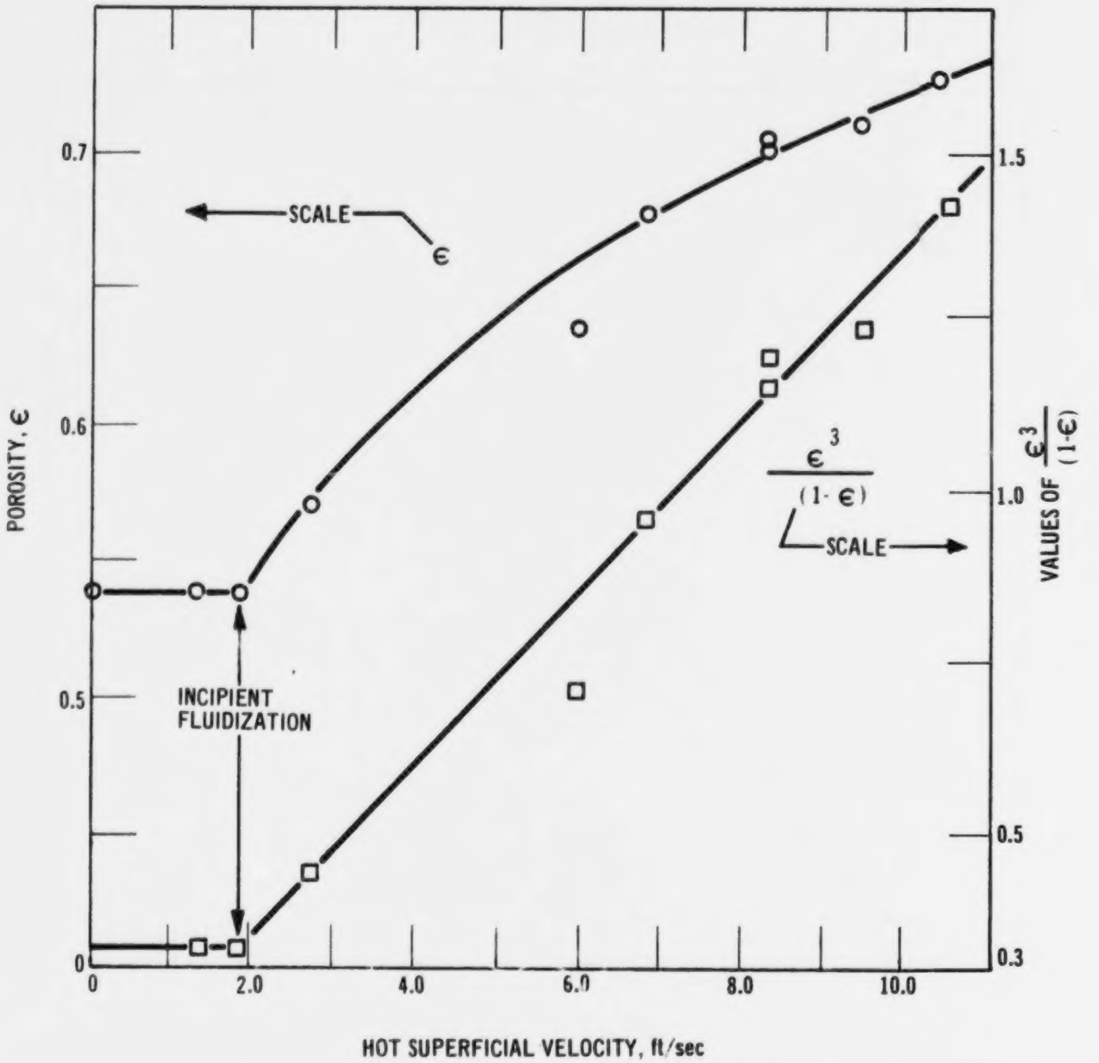

Figure 4. Variation of porosity ϵ (see equation 3) and ϵ^3 / $(1 - \epsilon)$ (see equation 2) with hot superficial velocity. The second group shows correlation between theory and experiment.

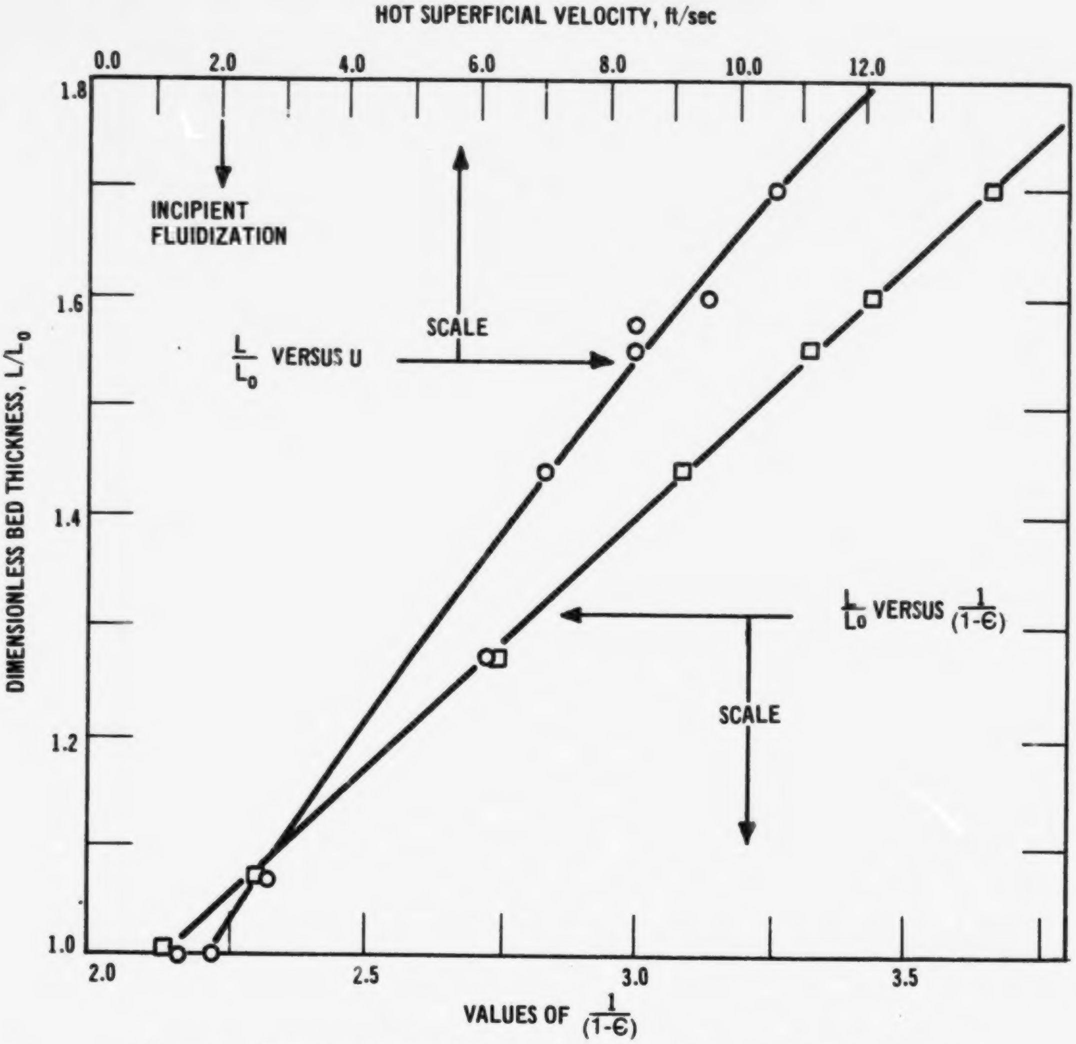

Figure 5. Variation of the dimensionless bed thickness L/Lo with both the superficial velocity U (curve calculated from equation 2 using experimental values of U_o and L_o) and with 1/(1-Є) as substantiation of equation 4.

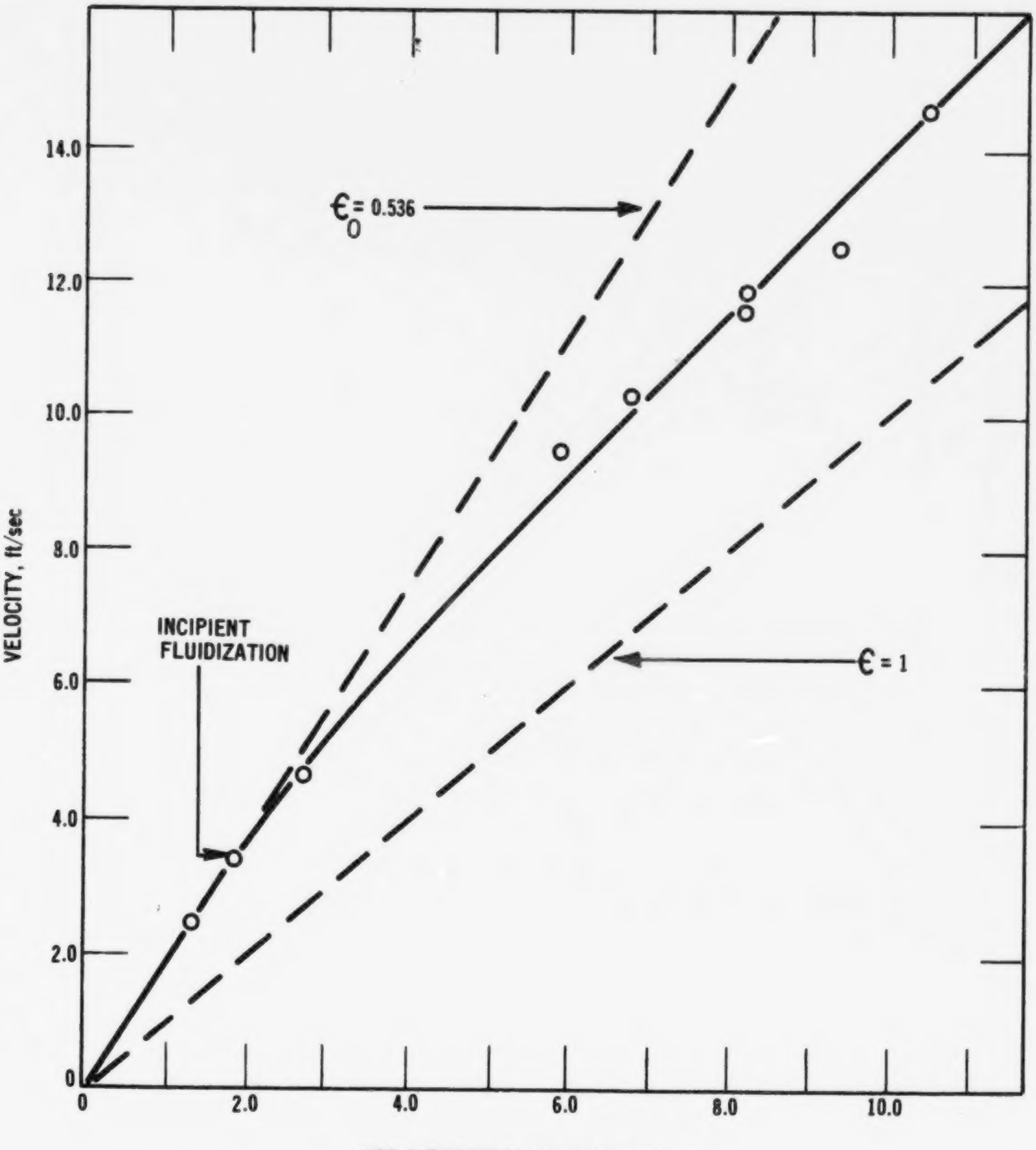

Figure 6. Variation of actual velocity with hot superficial velocity u. The dotted lines indicate what the velocity would be if the bed had a constant porosity equivalent to fixed bed (ϵ=0.536) or if no bed were present (ϵ=i).

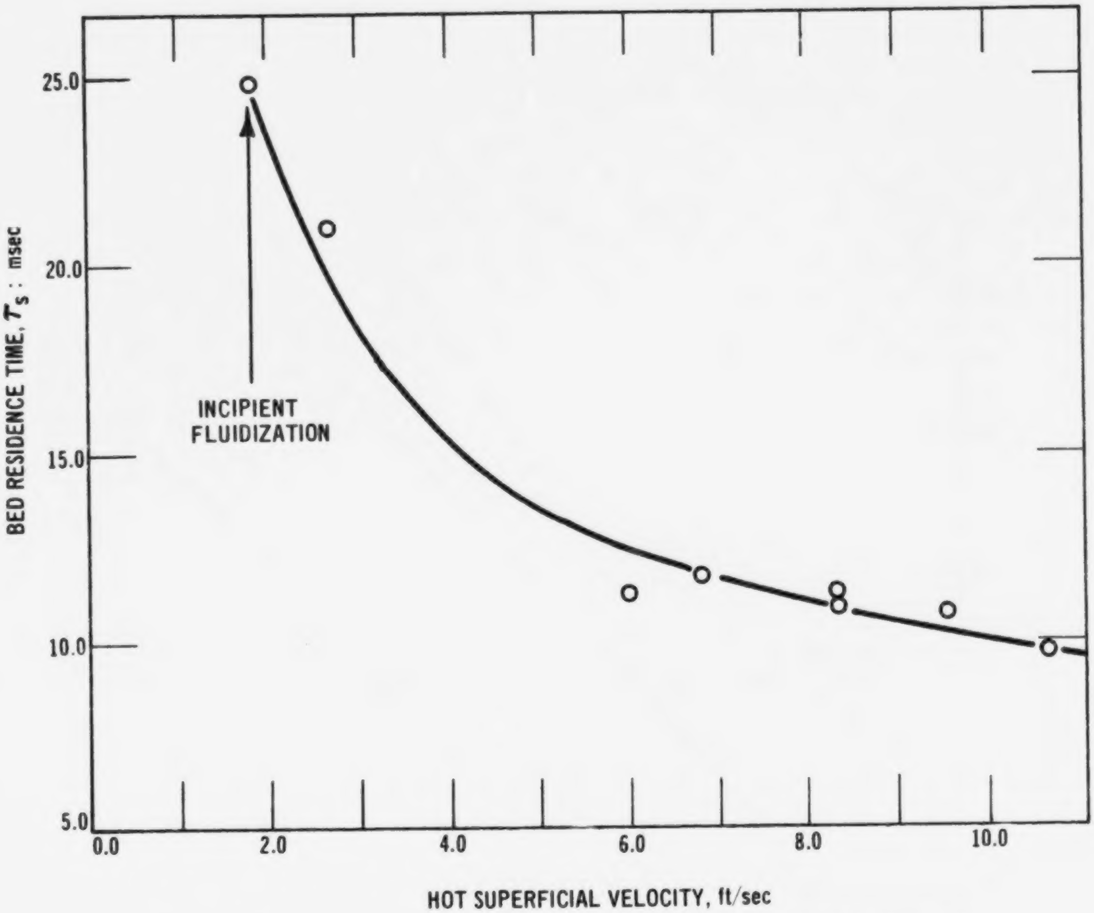

Figure 7. Variation of bed residence time τ_S with superficial velocity.

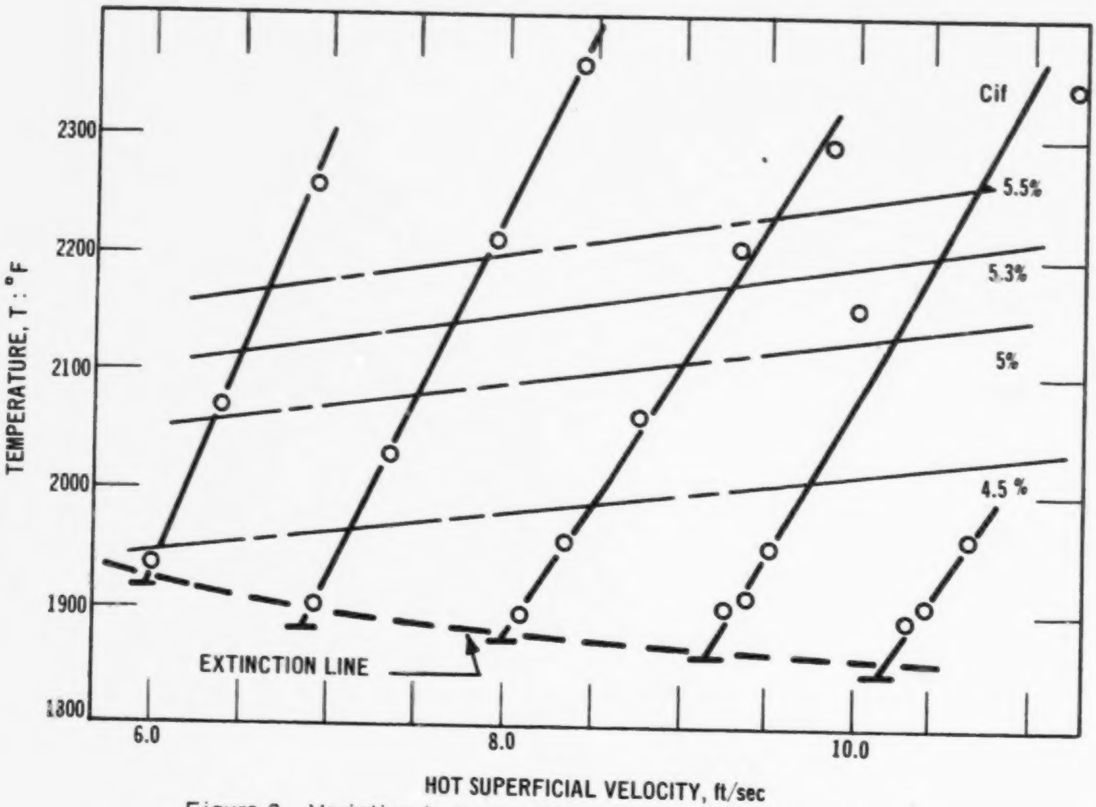

Figure 8. Variation in temperature with hot superficial velocity. The initial fuel concentration is also indicated. Extinction occurs at the low temperature and low initial fuel concentration.

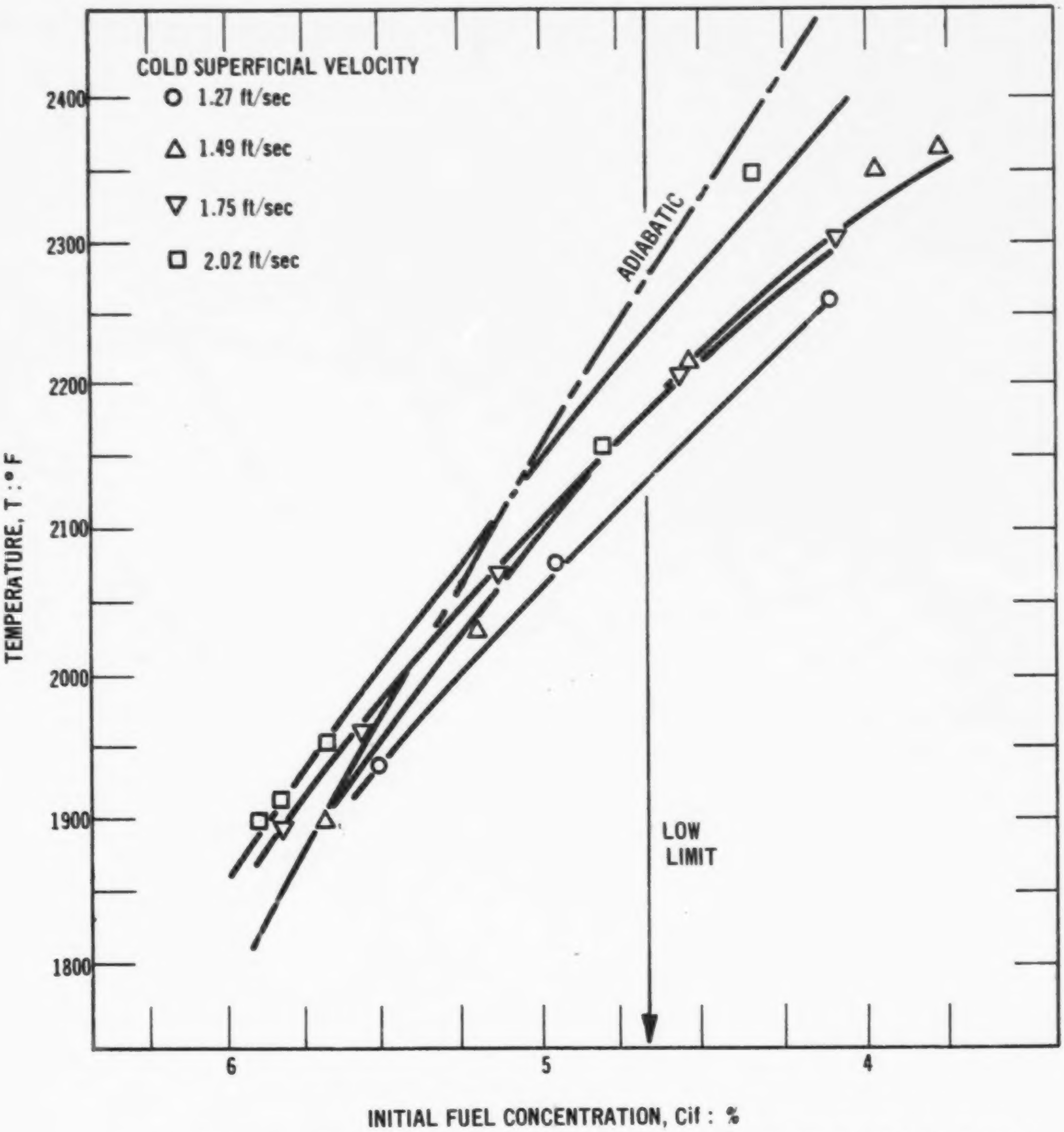

Figure 9. Temperature as function of initial fuel concentration for differing cold superficial velocities. Also indicated are the adiabatic line and the generally accepted lower flammability limit.

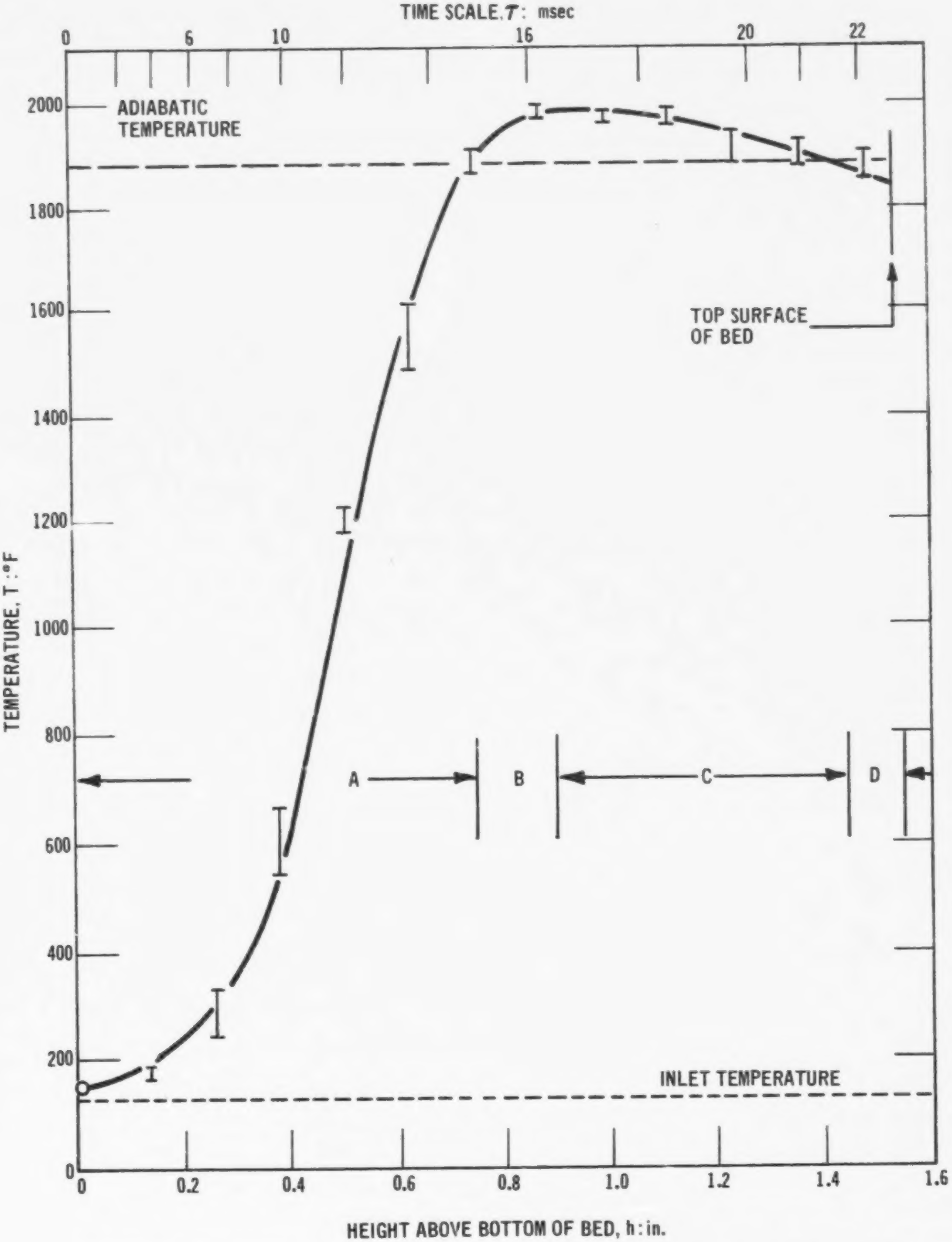

Figure 10. Temperature as a function of height in the bed with inlet and adiabatic temperatures indicated. An auxiliary axis shows the elapsed time in milliseconds.

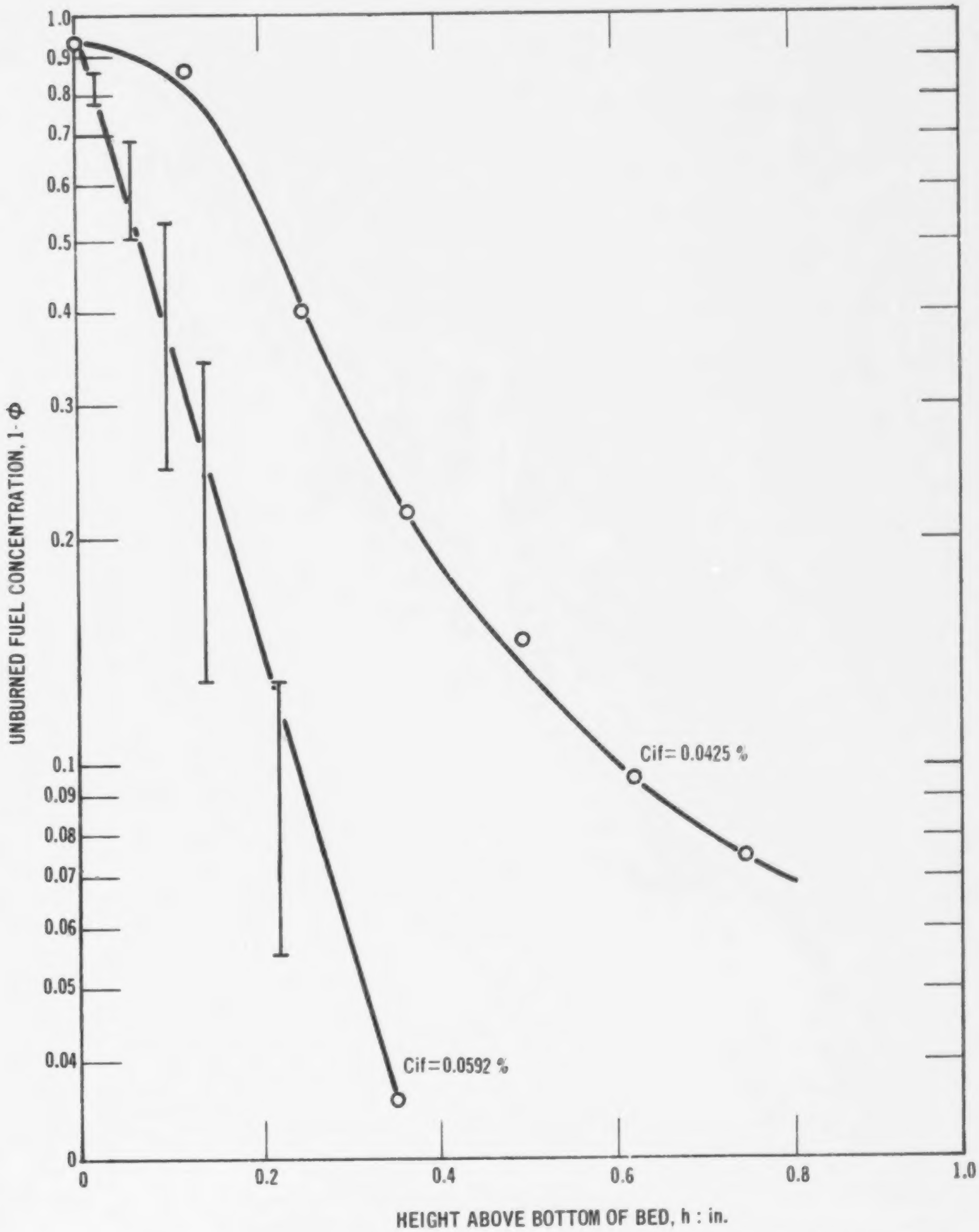

Figure 11. Unburned fuel concentration (calculated from CO_2 analysis) as a function of height in the bed for two initial fuel concentrations.

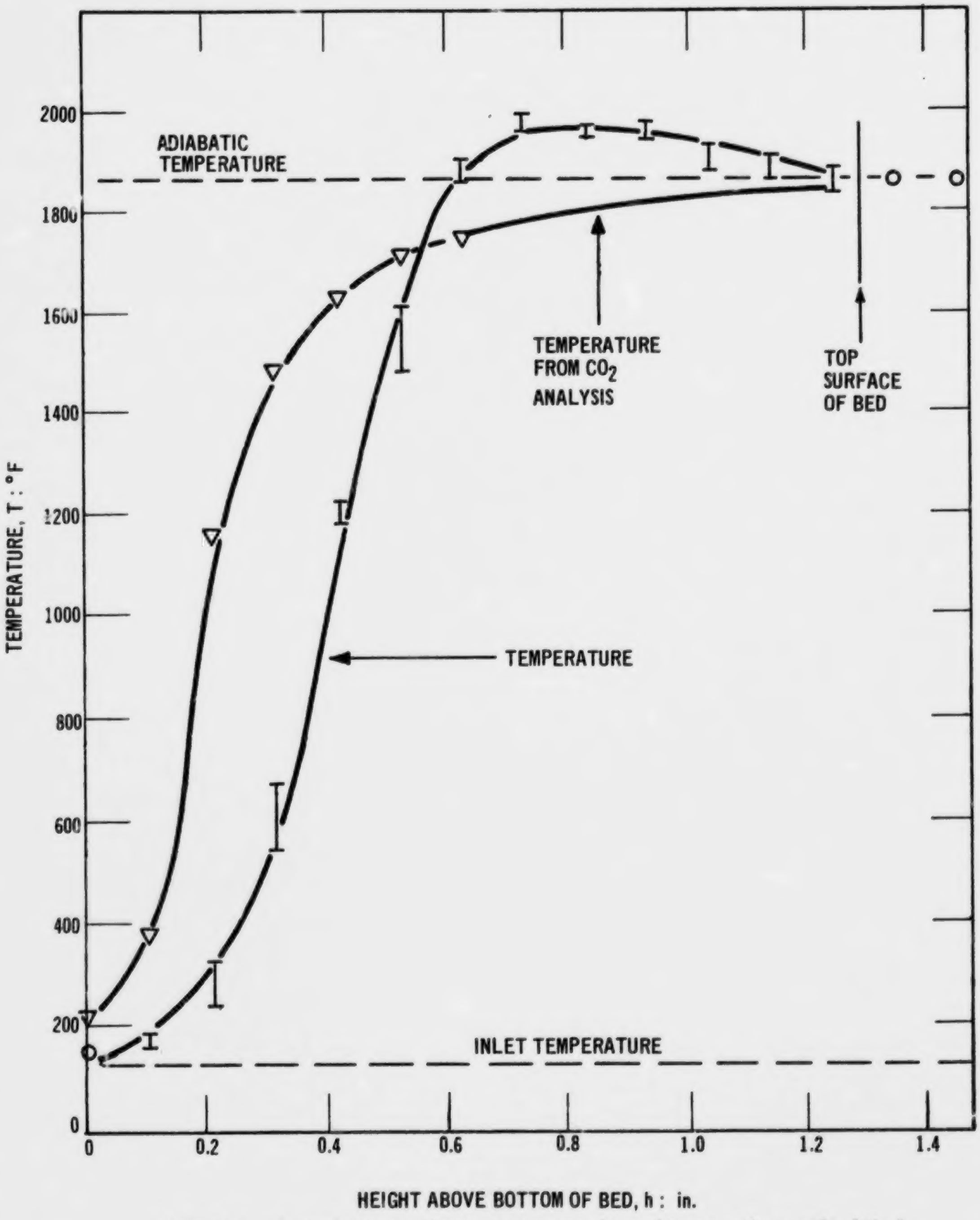

Figure 12. Actual temperature measurements and temperature calculated from the completeness of combustion as a function of height in the bed.

SESSION III:

Gasification/Desulfurization

SESSION CHAIRMAN:

Dr. E. Gorin, Consolidation Coal

1. FLUIDIZED-BED GASIFICATION—PROCESS AND EQUIPMENT DEVELOPMENT

J. T. STEWART AND E. K. DIEHL

Bituminous Coal Research, Inc.

INTRODUCTION

As part of its broad gas generator research and development program sponsored by the Office of Coal Research, U.S. Department of the Interior, Bituminous Coal Research, Inc., (BCR), is developing a multiple fluidized-bed coal gasification process for the production of low-Btu fuel gas. The goal of the multiple fluidized-bed system is the gasification of both caking and non-caking coal, with fuel gas being the only product.

Several government and industry co-sponsored coal gasification programs are at the pilot plant stage. These include the IGT Hygas process, the BCR BI-GAS process, and the Consolidation Coal Company's CO_2 acceptor process. Such processes are designed to generate high-Btu gas, i.e., gas having a heating value in excess of 900 Btu/scf. Gas of this quality can be used as a direct substitute for natural gas. Steam boiler and gas turbine applications for electrical power generation do not, however, need this high-Btu gas. Moreover, the optimum fuel gas heating value required for combined cycle applications can be of the order of 150 Btu/scf. The primary purpose of the multiple fluidized-bed coal gasification system is, then, the production of low-Btu fuel gas for the generation of electrical energy by means of the combined cycle.

This paper describes the BCR fluidized-bed gasifier concept and summarizes the work done by BCR in its development program. The program began with laboratory scale kinetic experiments and has progressed through the semi-continuous operation of a small fluidized-bed batch reactor.

With the aid of an engineering subcontractor, a process and equipment development unit (PEDU) has now been designed. The PEDU, to be located at the BCR Research Center at Monroeville, Pa., consists of three fluidized-bed reactors, a gas quenching and scrubbing system, facilities to preheat reactor inlet gases, and solids handling equipment. Designed to gasify 100 lb coal/hr, the PEDU will operate at 250 psia and at temperatures to 2100°F.

THREE STAGE FLUIDIZED BED CONCEPT

The goal of the multiple fluidized-bed system is the gasification of both caking and non-caking coal, with fuel gas being the only product. End use of the gasification product dictates the operating conditions as well as the gasifying medium. Thus, gasifying with air and steam will yield a low-Btu fuel gas; gasification with oxygen and steam can yield a higher Btu gas containing a greater proportion of combustible components; and gasification with carbon dioxide can yield a carbon monoxide-rich gas that could serve as a fuel for power generation by MHD.

A 3-stage system was chosen as one with the probable minimum number of stages necessary to meet the requirement of starting with any rank of coal and producing no tars or oils as a waste or by-product. Figure 1 is the design material balance for the proposed fluidized bed PEDU. Stage 1 receives raw coal and functions as the pretreatment step. The devolatilized coal flows by gravity to Stage 2 and then to Stage 3, which operates as the final carbon burn-up reactor. Several pretreatment mediums have been investigated by others and have been shown to be effective. These include air alone, and steam or carbon dioxide diluted with nitrogen and containing small amounts of air. In this scheme, Stage 3 flue gas is used as the fluidizing medium for Stage 1.

Stage 2 is the major gasification stage. The devolatilized coal is gasified with air and either steam or carbon dioxide to generate the desired product gas. In addition, Stage 1 flue gas is fed to Stage 2 where the entrained tars and oils are gasified. Stage 3 operates at the highest temperature and serves to maximize carbon utilization. The ash discharged from Stage 3 will contain a minimum amount of carbon. Hot flue gas from Stage 3 flows to Stage 1 and completes the cycle.

LABORATORY INVESTIGATIONS

The literature abounds with information regarding the kinetics of carbon/steam and carbon/carbon dioxide reactions, but there is often little agreement among the results of different investigators. For example, reported activation energies for the carbon/carbon dioxide reaction range from 25 to 90 kcal/mole. This wide variance may be attributed to the different types and ranks of carbon used, the different temperature and pressure ranges investigated, and the various simplifications and interpretations of the observed data. The different results are indicative of the different reaction mechanisms and rate controlling steps that occur under different experimental conditions. No correlations were found in the literature that could adequately describe the kinetics of the gasification reactions to the devolatilized coal or "char" that would be produced in the first step of the multiple fluidized-bed gasification scheme. Therefore, laboratory scale kinetic studies were begun.

Six widely different chars were chosen as the basis for the laboratory studies. The chars ranged in volatile content from 3 to 12 percent and were produced from all ranks of coal, from lignite to a highly caking Pittsburgh seam coal. Table 1 shows the chemical analyses of the selected chars.

EXPERIMENTAL EQUIPMENT AND PROCEDURE

A thermogravimetric balance was used to obtain the kinetic data. A schematic view of the TGA is shown in Figure 2.

Char is placed in the sample holder, a crucible or flat pan, which is connected to and suspended beneath the transducer coil and a precision spring. This entire assembly is mounted inside a quartz and Pyrex housing. During operation, the sample is located inside the well of the furnace where temperatures are continuously monitored by a chromel-alumel thermocouple.

Table 1. ANALYSIS OF CHARS USED IN REACTIVITY TESTS

Char sample number	Dry basis, %								
	Proximate			Ultimate					
	Volatile matter	Fixed carbon	Ash	Carbon	Hydrogen	Nitrogen	Sulfur	Ash	Oxygen
2455	2.85	88.9	7.65	88.7	0.8	1.3	0.6	7.7	0.9
2469	11.6	58.4	30.0	56.1	1.92	1.16	9.39	30.0	1.43
2927	2.1	74.6	22.5	73.5	1.35	0.62	2.89	22.7	0.0
1963	11.3	71.4	17.3	77.1	1.03	0.49	0.90	17.3	4.18
2280	5.9	82.5	11.6	82.0	1.8	-	0.37	11.6	-
2655	4.2	83.5	12.3	83.4	1.42	0.9	0.76	12.3	1.22

The char is brought to the chosen reaction temperature in an inert atmosphere. The inert gas stream is turned off; simultaneously the reaction gas stream is turned on. Sample weight loss is then recorded as a function of time. Data precision is checked and maintained within ± 0.1 percent weight loss/unit time by duplicating all experimental runs.

As the test progresses, changes in sample weight cause an extension or contraction of the spring which changes the positional relationship of the armature and transducer coil. A resulting electrical signal proportional to the change in sample weight is developed, amplified, and fed to the vertical (Y) axis of the recorder. The input to the horizontal (X) axis of the recorder is proportional to time or temperature.

MATHEMATICAL MODEL AND REGRESSION ANALYSIS

The purpose of this kinetic study is to establish the rate-controlling step and thus rate equations for the char/steam and char/carbon dioxide reactions. The study will also determine the effects of temperature, reacting gas concentration, and particle size.

The conversion of solids in a heterogeneous gas-solids reaction can follow one of two extremes. At one extreme the diffusion of gaseous reactant into the particle is rapid enough, compared to the chemical reaction rate, that the reaction takes place at the same time and at the same rate everywhere. This is called the continuous reaction model. If diffusion into the particle is slow, the reaction is restricted to a thin shell which moves from the outside of the particle inward. This is the unreacted core model with diffusion controlling.

The appropriate model may be chosen by determining the time needed for complete conversion of solids of different sizes, as summarized below:

The parameter t_c, defined as the time needed for complete conversion is:

1. Independent of particle diameter for the continuous reaction model.
2. Directly proportional to particle diameter for the unreacted core model, with chemical reaction at the reaction front as the rate controlling step.
3. Directly proportional to the square of the particle diameter for the unreacted core model, with diffusion through the ash layer as the rate controlling step.

Experiments were carried out with particles ranging in size from 70 mesh (210μm) to minus 325 mesh, (44μm). Within the temperature ranges investigated, the reaction rate was independent of particle diameter. Therefore, the continuous reaction model was chosen to develop a rate equation, as follows: the rate of carbon is proportional to the concentration of reacting gas around particles times the amount of carbon left unreacted. In terms of the fraction of carbon reacted, X, this becomes

$$\frac{dX}{dt} = kC^n(1 - X) \qquad (1)$$

where: C^n is the concentration of reacting gas to some power, n.

Rearranging equation (1) gives:

$$\frac{dX}{(1 - X)} = kC^n dt \qquad (2)$$

Integrating equation (2) yields:

$$\ln(1 - X) = -kC^n t$$
$$\text{or}$$
$$(1 - X) = e^{-kC^n t} \qquad (3)$$

The proportionality or rate constant, k, may be assumed to vary with temperature in an Arrhenian fashion:

$$k = ae^{\frac{-E}{RT}} \qquad (4)$$

Combining equations (3) and (4) gives:

$$(1 - X) = e^{-ae^{\frac{-E}{RT}} C^n t}$$

Letting X now represent the amount of char reacted, the complete rate equation becomes:

$$(1 - X) = Ash + (1 - Ash)\, e^{-ae^{-\frac{E}{RT}} C^{n} t} \tag{5}$$

where:

X = fraction of char reacted

T = temperature, °K

G = concentration of reacting gas

t = time, minutes

k = apparent reactivity, $\frac{\text{lb C reacted}}{\text{lb C inventory min}}$

Ash = weight percent of ash in unreacted char

E = activation energy, $\frac{\text{cal}}{\text{mole}}$

R = the gas constant = 1.987 $\frac{\text{cal}}{\text{mole }^{\circ}\text{K}}$

n = the order of reaction with respect to reacting gas

a = the Arrhenius constant (frequency factor), min^{-1}

The experimental data are in the form of a plot of (1 - X) as a function of t. The constants a, E, and n may then be found from equation (5) when (1 - X), Ash, T, C, and t are known variables. The regression analysis proceeds in two steps. For each value of Ash, C, and T, (1 - X) versus t is solved for the constant M as follows.

Let $M = ae^{-E/RT} C^{n}$

then $$(1 - X) = Ash + (1 - Ash)\, e^{Mt} \tag{6}$$

and $$\ln \frac{(1 - X - Ash)}{(1 - Ash)} = Mt \tag{7}$$

The square of the error in this equality is:

$$\varepsilon = \Sigma\, [\ln \left(\frac{1\text{-}Xi\text{-}Ash}{1 - Ash}\right) - Mt_i^{\,2}]^2 \tag{8}$$

where: ε is the error and the subscript i denotes individual data points. ε is minimized by differentiating and setting it equal to zero.

$$\frac{d\varepsilon}{dM} = O = -2\,\Sigma\, [t_i \ln \left(\frac{1\text{-}X_i\text{-}Ash}{1 - Ash}\right) - Mt_i^{\,2}]$$

$$O = \Sigma\, [t_i \ln \left(\frac{1\text{-}X_i\text{-}Ash}{1 - Ash}\right) - Mt_i^{\,2}]$$

$$M = \frac{\Sigma\, t_i \ln \left(\frac{1\text{-}X_i\text{-}Ash}{1 - Ash}\right)}{\Sigma t_i^{\,2}}$$

$$M = \frac{\Sigma\, t_i \ln(1\text{-}X\text{-}Ash) - \ln(1\text{-}Ash)\, \Sigma t_i}{\Sigma t_i^{\,2}} \tag{9}$$

Once M is found from the above equation for each C and T, a multiple linear regression approach may be used to find the constants -E/R, a, and n by solving

$$M = ae^{-E/RT} (C^{n})$$

or $$\ln M = \ln(a - E/RT - n) \ln C \tag{10}$$

Again, the partial derivatives of the error function with respect to M, T, and C are set equal to zero; the three resulting equations are solved simultaneously for a, -E/R, and n.

EXPERIMENTAL RESULTS

A series of photomicrographs gave the first physical clues leading to the selection of a reaction model. Figures 3 through 6 show unreacted char, 50 percent reacted char, 80 percent reacted char, and ash. It is immediately apparent that the average particle diameter does not change with increasing carbon burn-off. The particles become increasingly porous, but even the ash residue retains a skeletal structure similar in overall dimensions to the unreacted char. Along with these physical observations, the experimental data showed that the time needed for complete reaction was independent of particle diameter. Rate equations were thus developed for the six chars. For example, the rate equation for the reaction of char No. 2455 with steam is:

$$(1\text{-}x) = Ash + (1\text{-}Ash)\, e^{-k (C_{H_2O})^{0.58}\, t}$$

$$k = 9.5 \times 10^4\, e^{-1.7 \times 10^4/T}$$

The significance of this rate equation is demonstrated in Figures 7 and 8. The activation energies are of the order of 40 kcal/mole. Both the char-steam and the char-carbon dioxide were found to be approximately half order with respect to reacting gas concentration.

The char-oxygen reaction was also studied. Within the temperature range of importance to this investigation, the reaction was controlled by mass transfer from the gas phase to the surface of the particle. Again the particles do not shrink as the reaction proceeds. Diffusion of reactants and products into and out of these small, porous chars is so fast compared to the chemical reaction step that the reaction may be thought of as taking place continuously throughout the particle.

The next step in the laboratory studies was the construction and operation of a small fluidized-bed batch reactor. Figure 9 is a schematic diagram of this system, and Figure 10 shows the actual equipment. The reactor was made from 1-1/2 in. schedule 40, type 310 stainless steel pipe. External heating was provided for operation at a maximum reactor temperature of 2200°F. The system is designed for operation at pressures to 10 atm.

Table 2. MATERIAL BALANCE FOR FLUIDIZED-BED BATCH REACTOR AIR-BLOWN GASIFICATION TEST NUMBER 4[a]

Component	Feed		Product	
	Mole, %	g moles/min	Mole, %	g moles/min
Oxygen	12.4	0.00846	0.12	0.0001
Nitrogen	46.4	0.03160	30.72	0.0316
Carbon dioxide	41.2	0.02824	21.45	0.0221
Carbon monoxide	0	0	47.71	0.0491
Hydrogen	0	0	0	0
TOTAL	100.0	0.06830		0.1029

Feed gas rate: 1530 sml/min
Product gas rate: 2300 sml/min

	In	Out
Total g moles Carbon	0.02824	0.0712
Total g moles Oxygen	0.03770	0.0467
Total g moles Nitrogen	0.03160	0.03160

[a] Carbon gasification rate, 0.0429 g moles/min
Carbon Dioxide utilization, 21.7 percent
Reactor Pressure, 68 psia
Reactor Temperature, 940°C
Initial Charge to Reactor, 20 g char, BCR Lot 2455

The batch reactor tests had a threefold purpose:

1. To verify the proposed rate equations.

2. To determine the degree of steam or carbon dioxide decomposition that could be achieved in a reactor of reasonable size.

3. To provide physical data such as minimum fluidizing velocity, attrition rate, elutriation losses, etc.

Table 2 is a material balance from a typical batch reactor test. The gasifying medium was air and carbon dioxide. At the reaction temperature of 940°C, the gasification rate of 0.0429 g moles/min was consistent with the predicted value of 0.0519 g moles/min. The product gas had a gross heating value of approximately 153 Btu/scf.

The success of the laboratory studies led to the design of the 100 lb/hr process and equipment development unit.

PROCESS AND EQUIPMENT DEVELOPMENT UNIT (PEDU)

The purpose of the PEDU is to provide the necessary parameters for the design and operation of a pilot scale or larger unit to demonstrate the process and the economic feasibility of fluidized-bed gasification for the commercial production of low-Btu fuel gas. The PEDU was designed to conform with the desire of the Office of Coal Research to have a flexible system designed with a nominal capacity of 100 lb coal/hr. Feed to the unit can be either coal or char with air or oxygen, or a mixture, as the oxidant and steam and/or carbon dioxide as the moderator.

Figure 11 is a schematic diagram showing the major process equipment. Coal is metered from the pressurized lock hopper through a rotary air-lock feeder and flows by gravity into the first reactor. Stage 1, the smallest reactor, has a reaction zone inside diameter of 10 inches and a disengaging zone of 16 inches. Stage 2 is the largest reactor with a reaction zone inside diameter of 16 inches and a 24-in. disengaging zone. The reaction zone diameter of Stage 3 is 12 inches with a 16-inch disengaging zone. All three reactors are approximately 11 feet high.

Refractory-lined cyclones are provided for stages 2 and 3 to recycle entrained solids to the bed. Solids are scrubbed from the product gas stream in a venturi scrubber, and the gas flows through iron oxide boxes for hydrogen sulfide removal and thence to a thermal oxidizer for disposal.

The next step in the development program is the construction and operation of the PEDU. Detail engineering, procurement, and erection will take approximately 12 months. A definitive cost estimate is currently being prepared for this phase of the program. The Office of Coal Research will then decide to what extent and in what manner this project will continue.

ACKNOWLEDGMENT

This paper is based on work carried out at Bituminous Coal Research, Inc., with support from the Office of Coal Research, U.S. Department of the Interior, under Contract No. 14-32-0001-1207.

Stream number	1		2		3		4		5		6	7		8		9	10	11
	Coal feed		Char from stage 1		Stage 1 flue gas		Stage 3 flue gas		Product gas		Air to stage 2	Char from stage 2		Char from stage 3		Air to stage 3	Steam to stage 3	Steam to stage 2
	lb	wt %	lb	wt %	moles	vol %	moles	vol %	moles	vol %	moles	lb	wt %	lb	wt %	moles	moles	moles
Coal (ash free)	93.8	93.8	72.6	92.1								26.4	81	3.6	37			
Ash	6.2	6.2	6.2	7.9								6.2	19	6.2	63			
H_2					0.95	13.7	0.95	16.2	4.7	21.3								
CO					1.53	22.0	1.53	26.0	5.16	23.4								
CO_2					0.37	5.3	0.37	6.3	1.04	4.7								
H_2O					0.55	7.9	0.55	9.4	2.00	9.0								
N_2					2.48	35.7	2.48	42.1	9.14	41.4								
H_2S									0.05	0.2								
Stage 1 off gas					1.06	15.4												
Total	100.0	100.0	78.8	100.0	6.94	100.0	5.88	100.0	22.1	100.0	8.43	32.6	100	9.8	100	3.14	1.50	2.5
Average mole wt					23.3		23.9		22.3		29					29	18	18
Temperature, °F	77		1200		1200		2100		2000		1000	2000		2100		1000	1000	1000
Pressure, psia					250		250		250		300	250				300	300	300
scfm					43.8		37.1		140.0		53.0					19.8	9.5	15.8
acfm					8.2		10.7		38.9		7.3					2.7	1.31	2.17

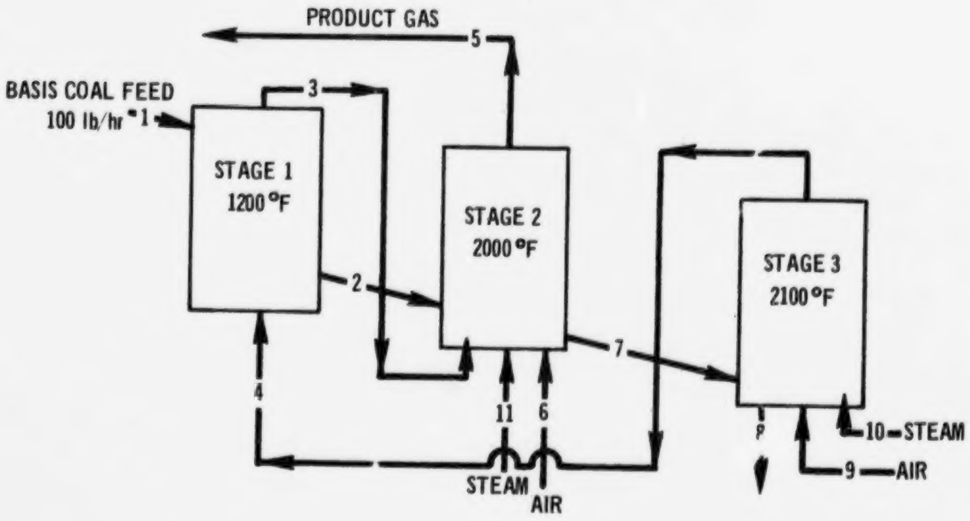

Figure 1. Material balance for gasification with air and steam.

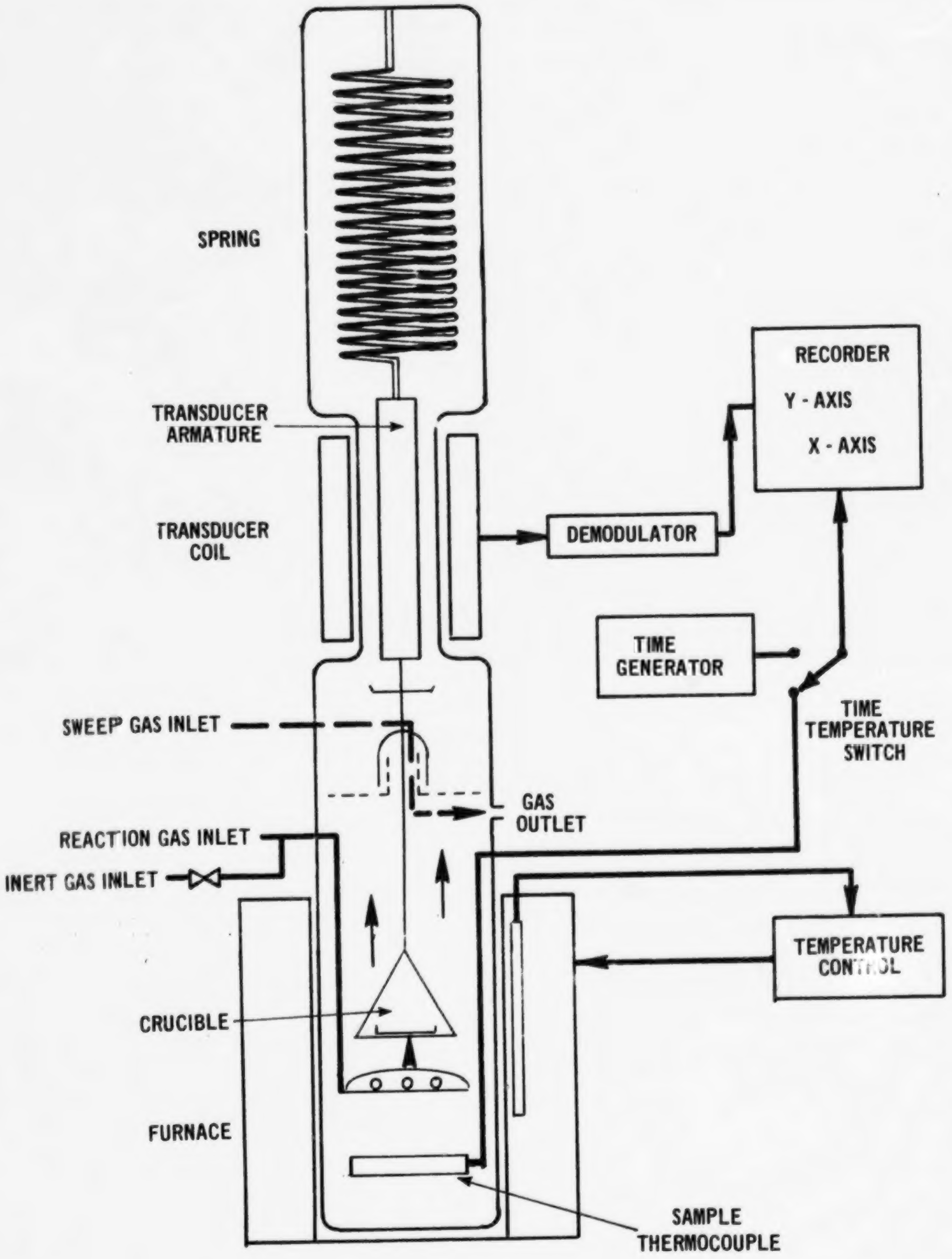

Figure 2. Schematic view of thermogravimetric analysis equipment.

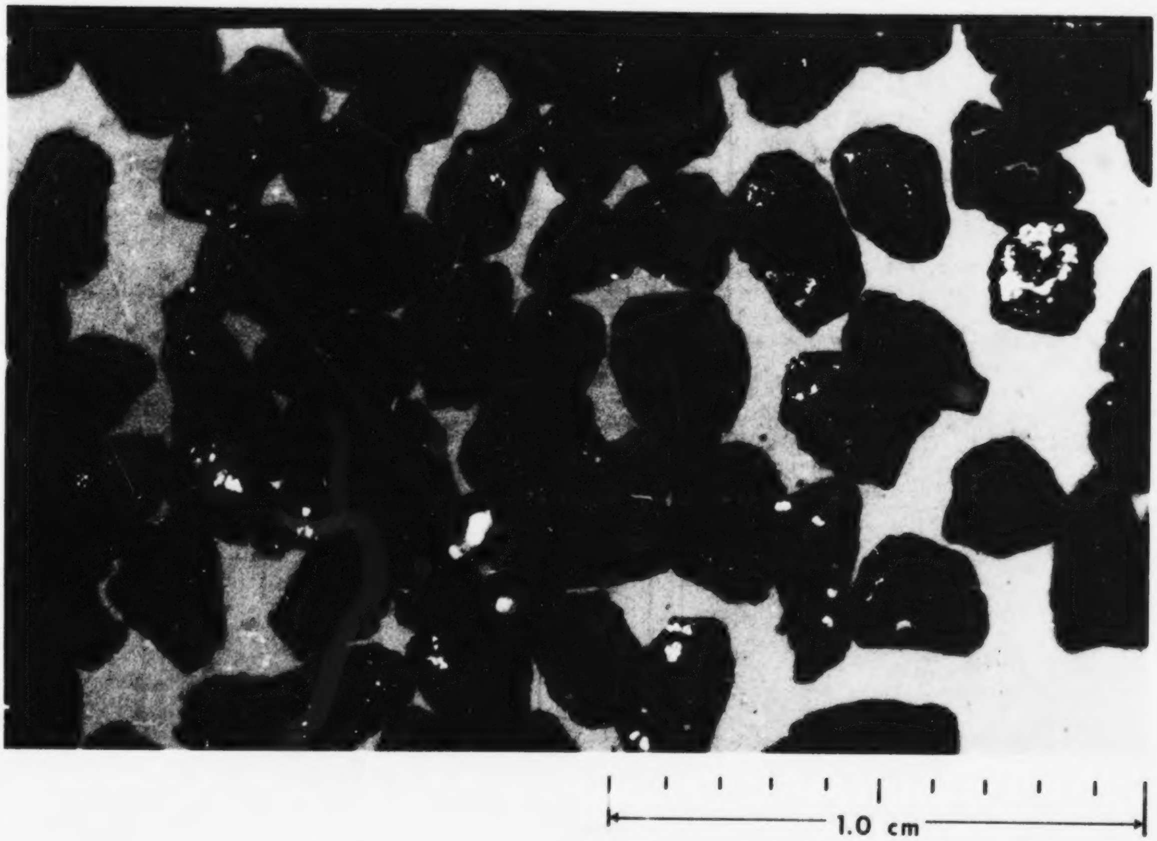

Figure 3. Photomicrograph of unreacted char.

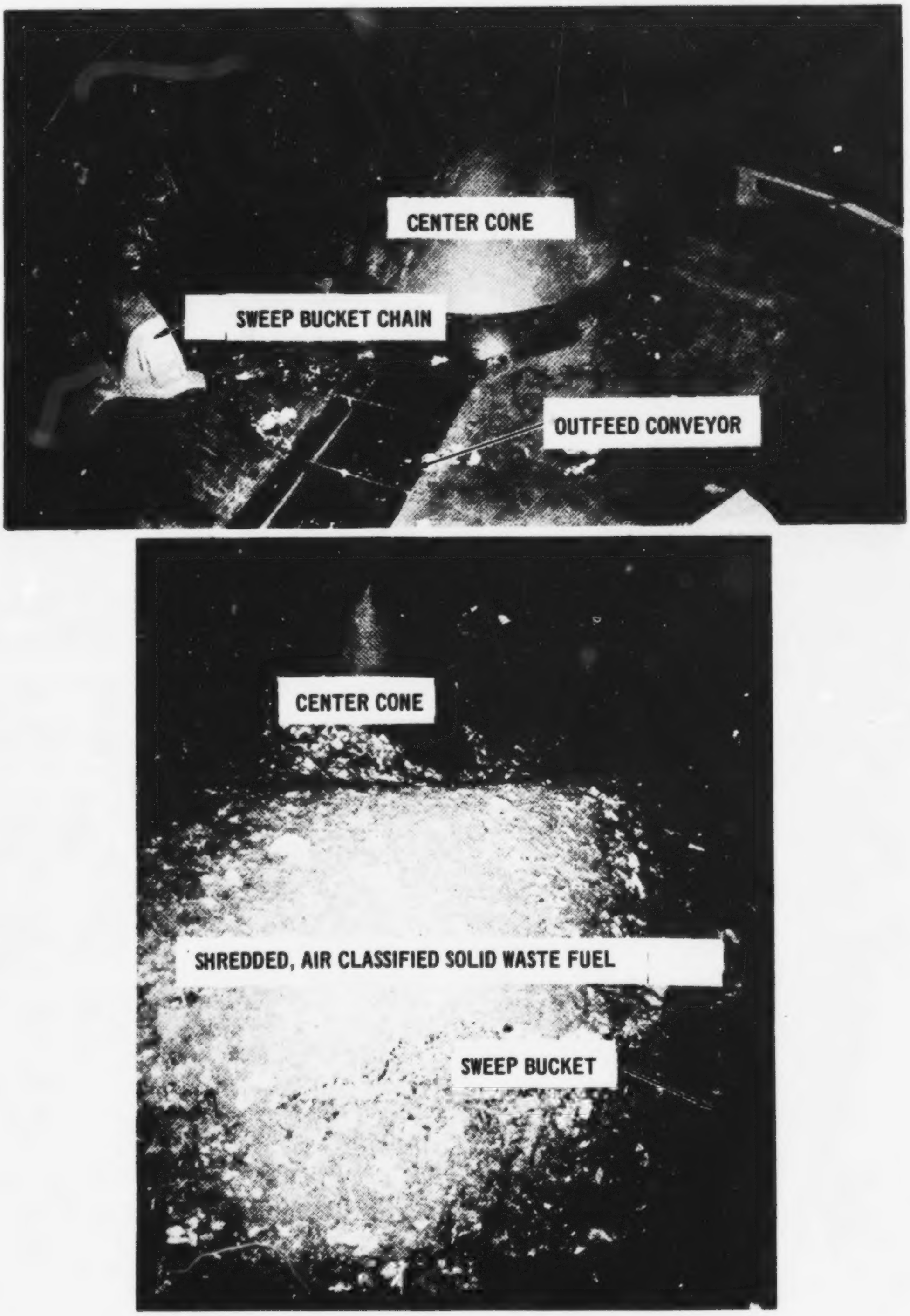

Figure 4. Photographic views of the solid waste storage tank interior.

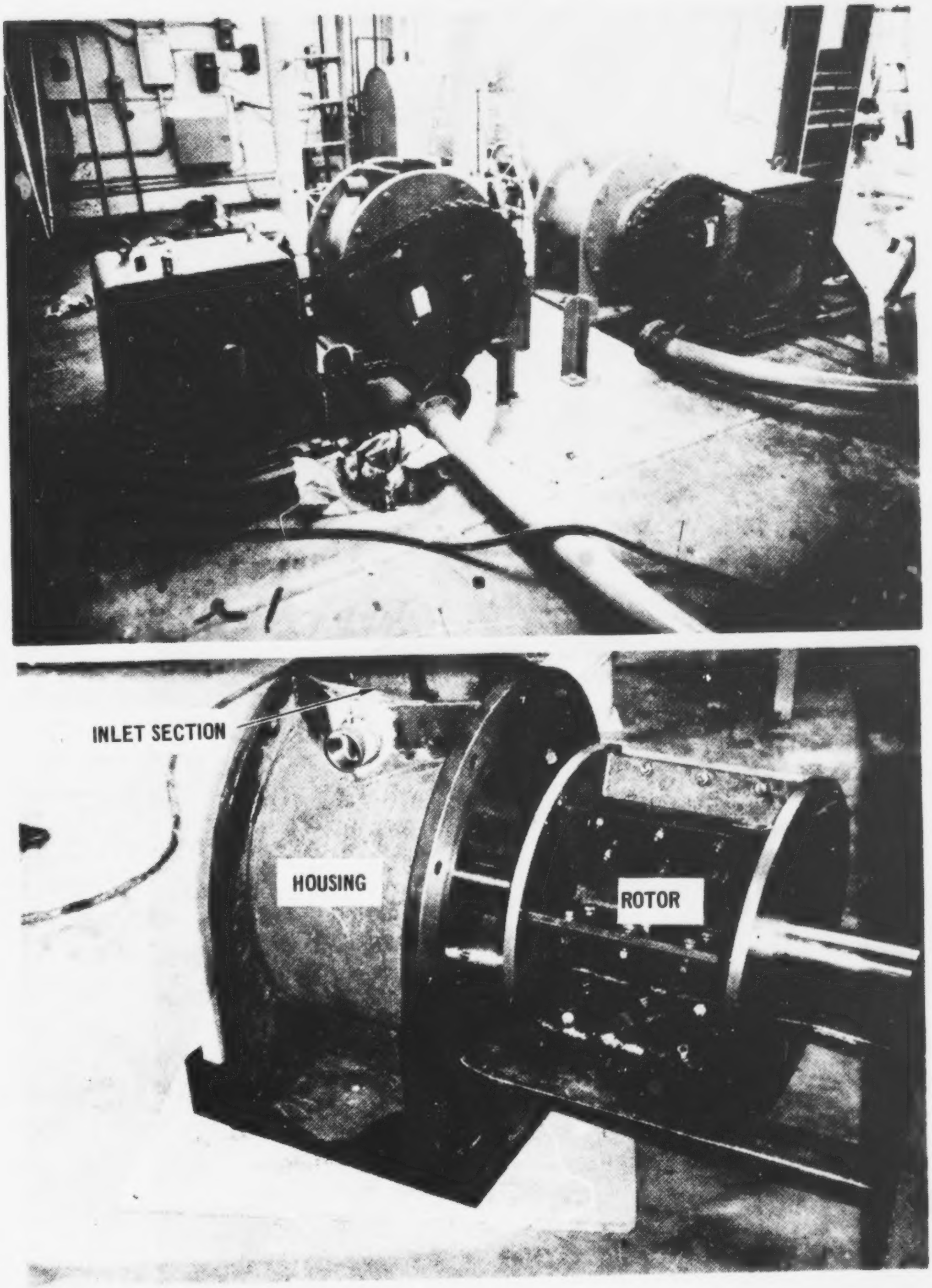

Figure 5. Airlock feeder valve.

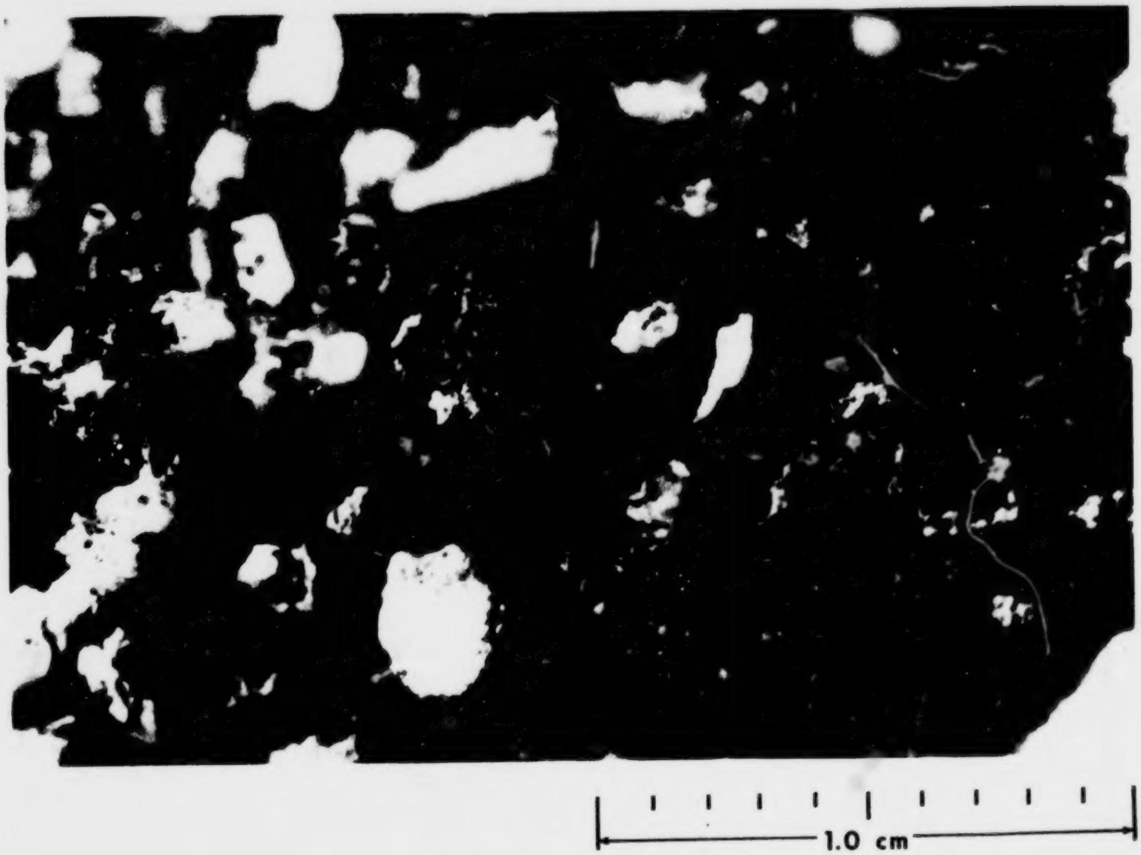

Figure 6. Photomicrograph of char ash (100% burn-off).

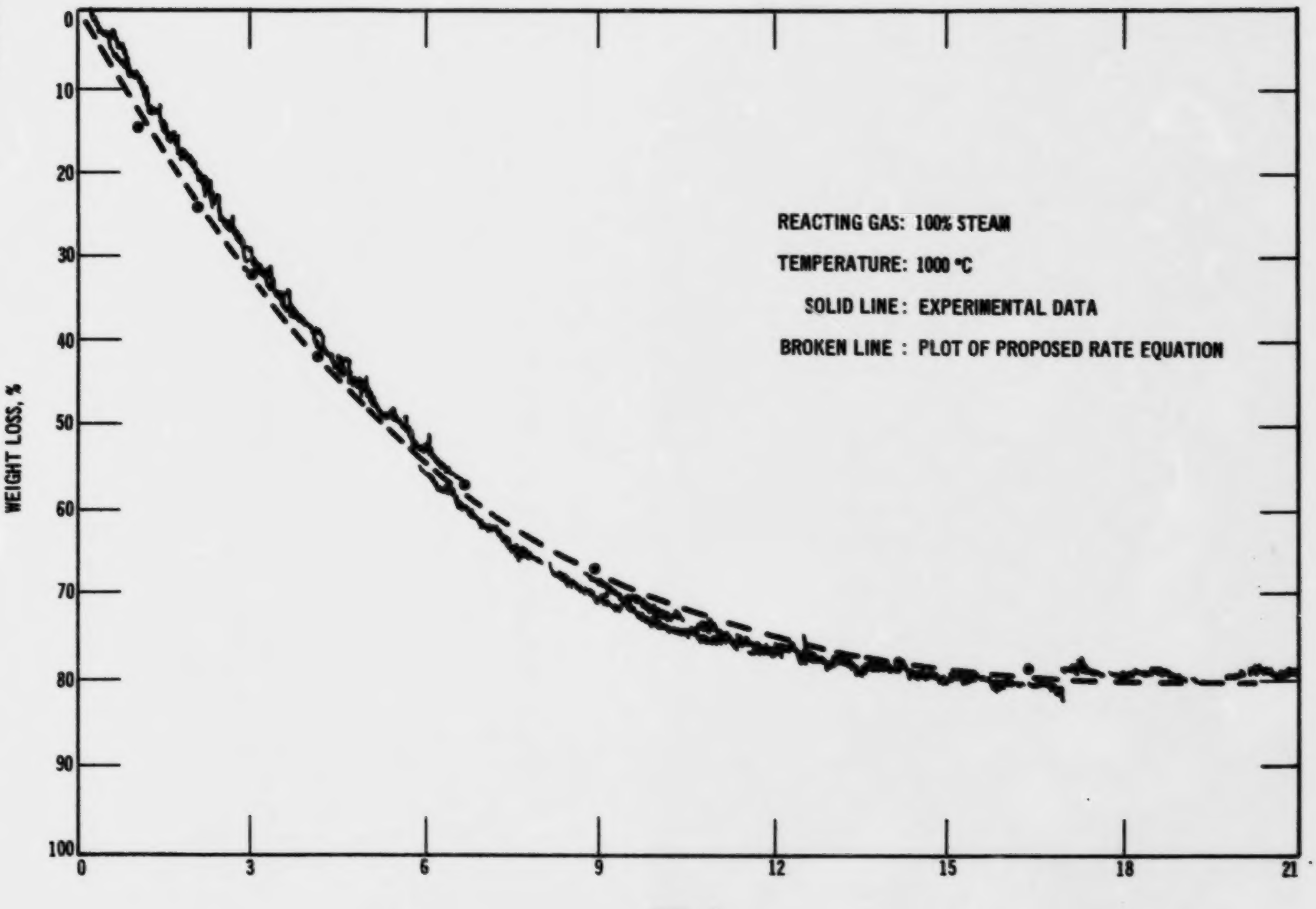

Figure 7. Typical correlation of reactivity data.

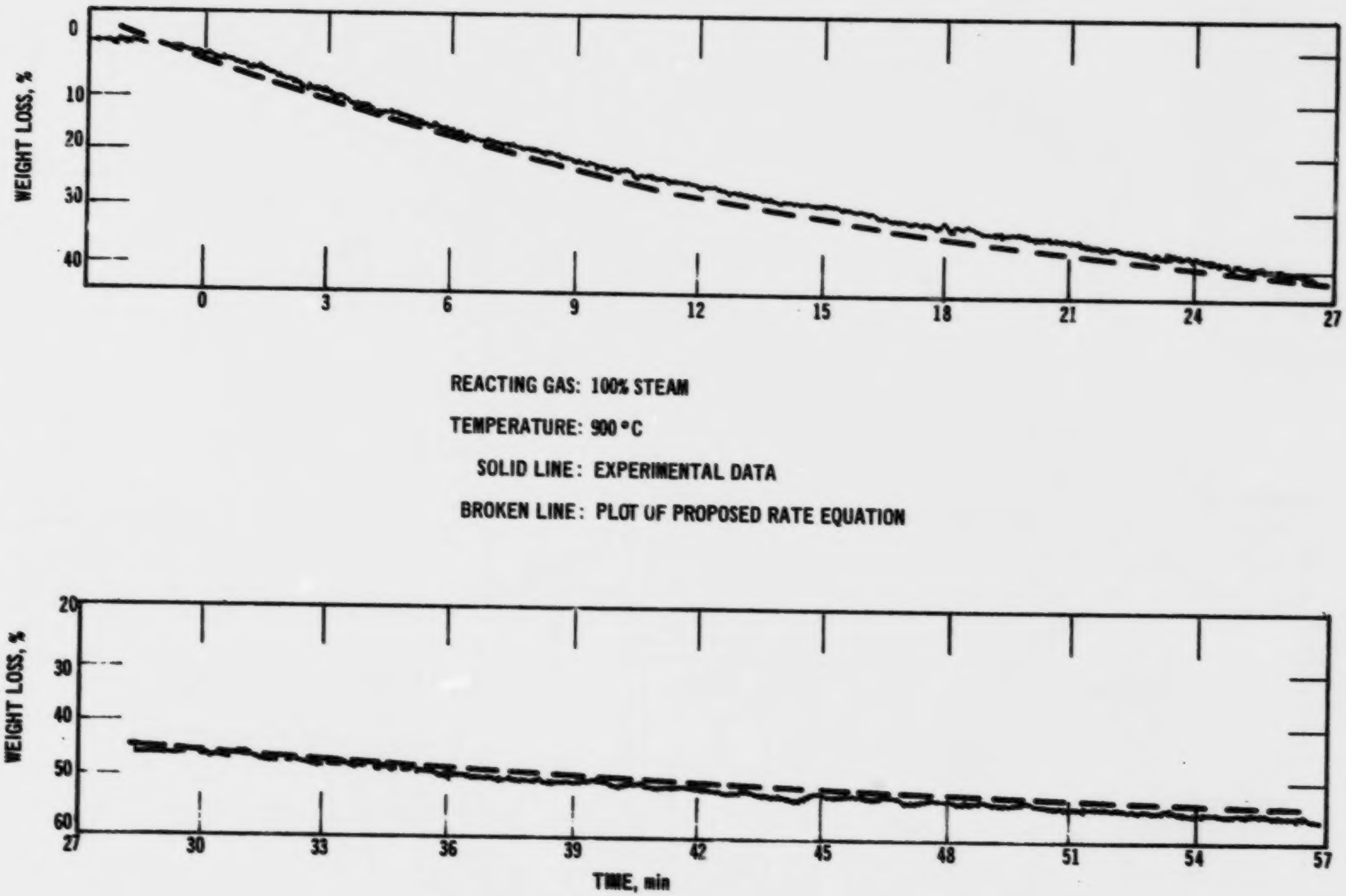

Figure 8. Typical correlation of char reactivity data.

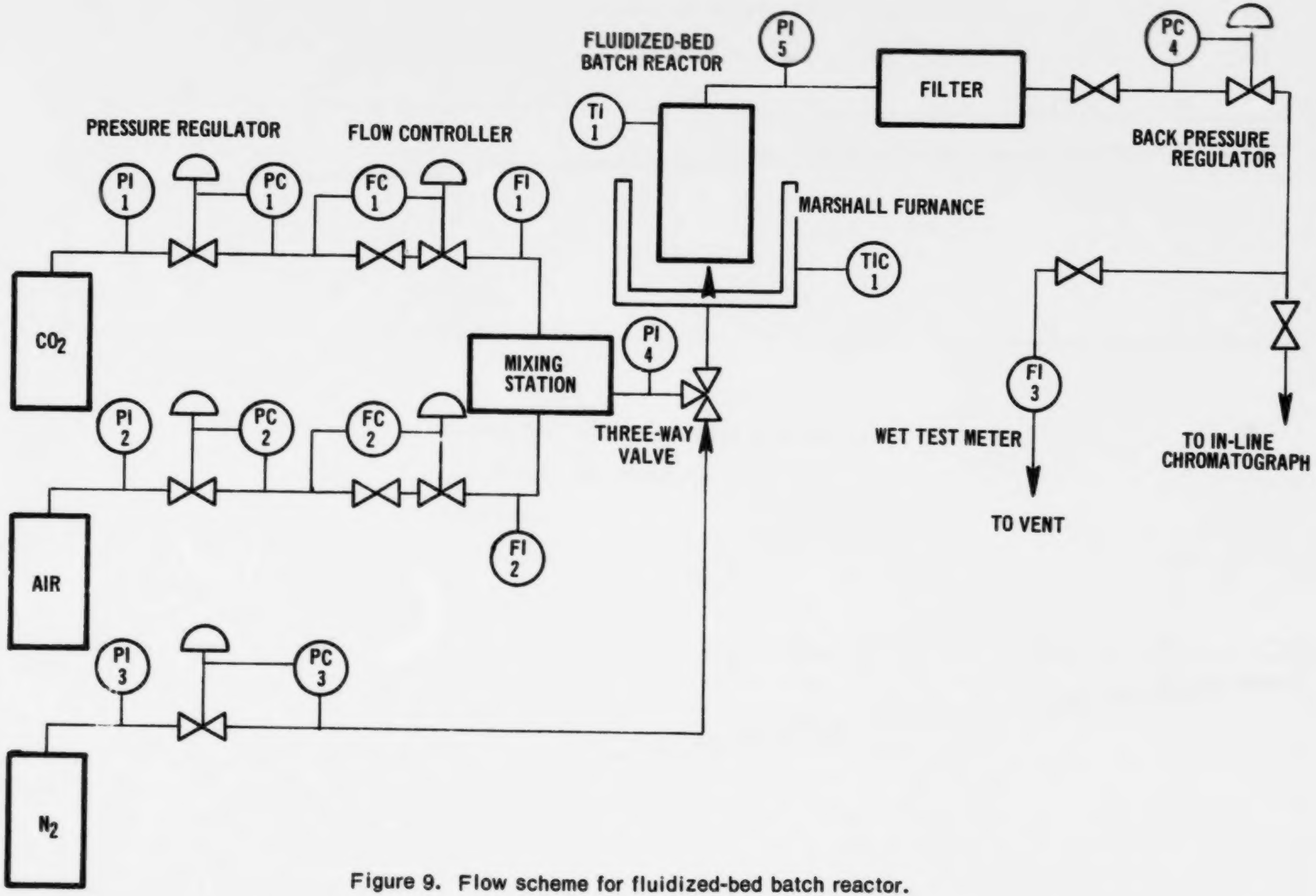

Figure 9. Flow scheme for fluidized-bed batch reactor.

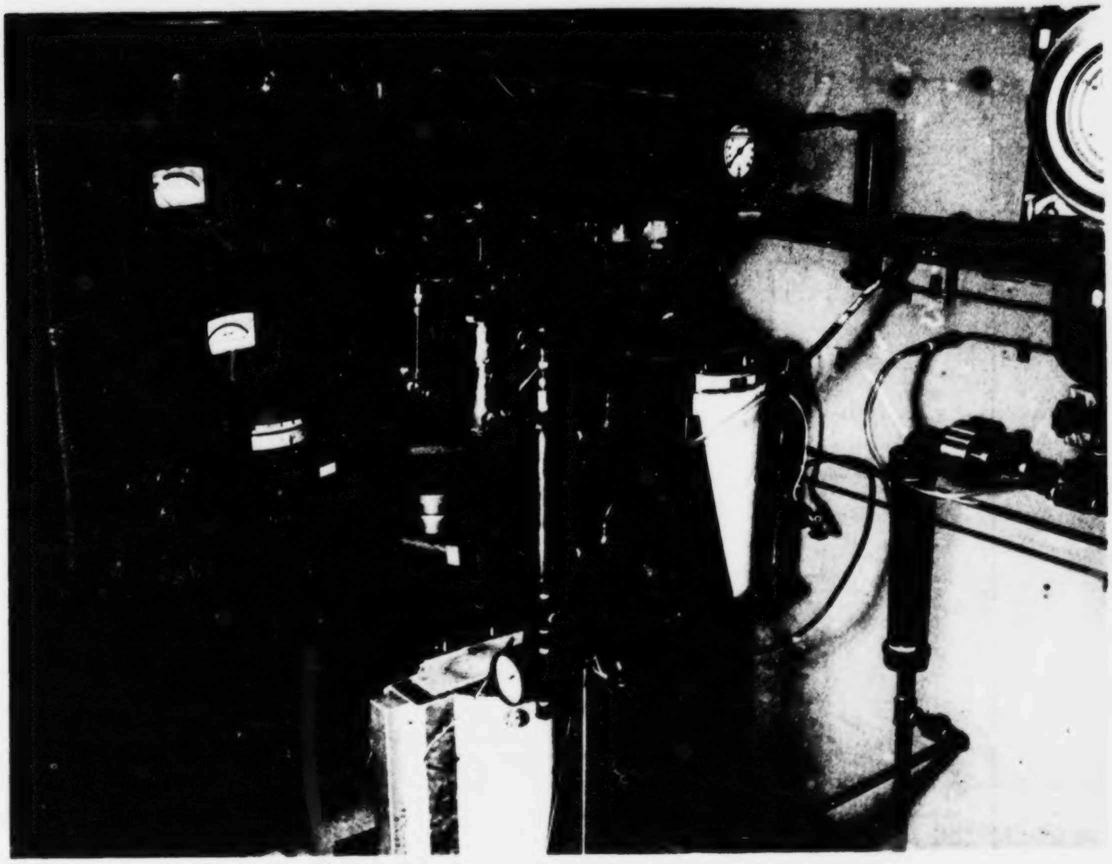

Figure 10. Fluidized-bed batch reactor system.

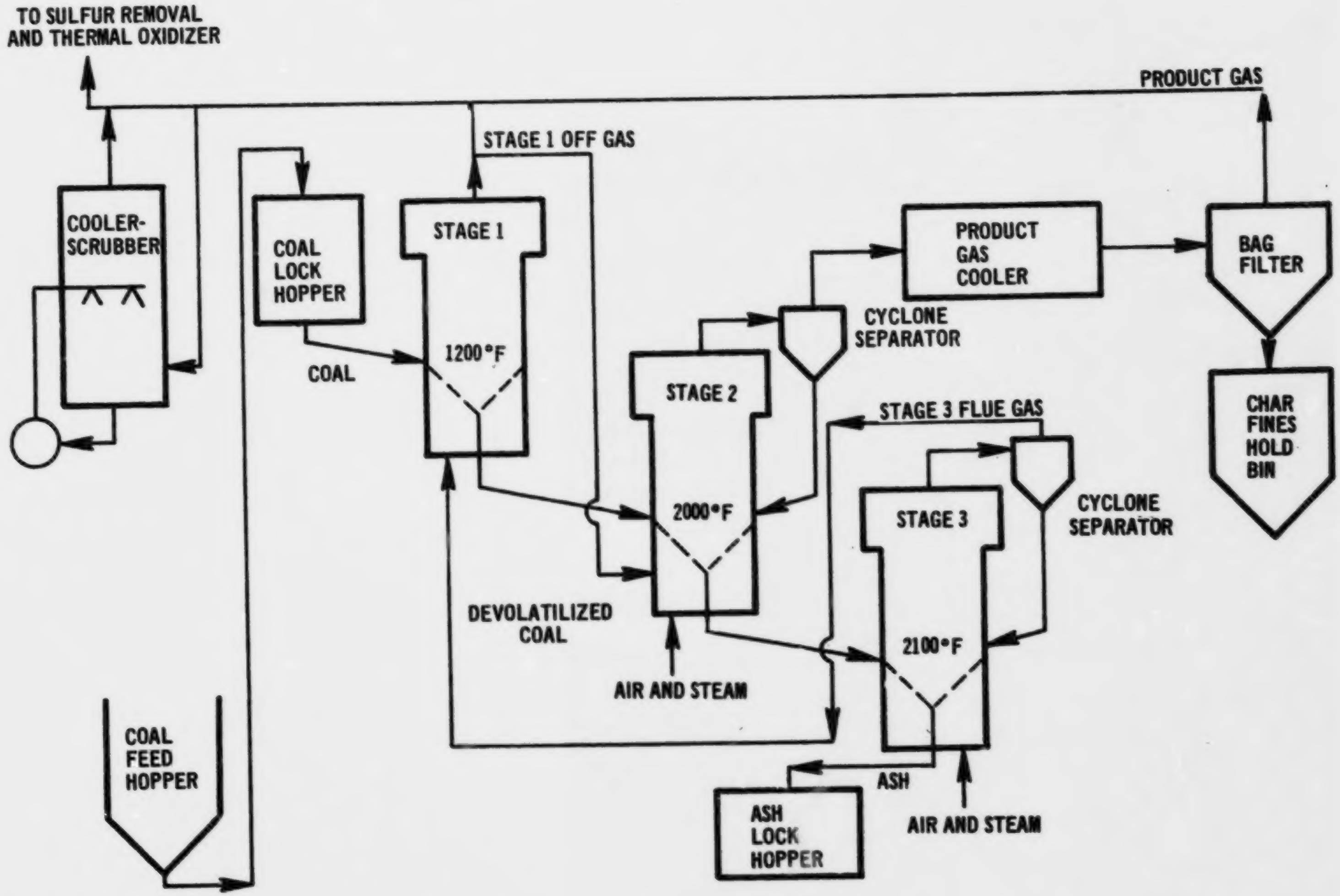

Figure 11. Fluidized-bed gasification PEDU process flow diagram.

2. HOT SULFUR REMOVAL FROM PRODUCER GAS

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U.S. Bureau of Mines
U. S. Department of the Interior

ABSTRACT

Sulfur-free gas for power generation or catalytic conversion to pipeline gas is needed to meet near term energy and antipollution requirements. Gasification of coal with air or oxygen and steam at elevated pressures supplies the gas, but cleaning is required to remove sulfur and particulate matter. A stirred-bed pressurized producer is described, and results are discussed for caking coals. Progress is reported in developing a process using a regenerable solid sorbent for removing hydrogen sulfide from hot producer gas with recovery of elemental sulfur formed during regeneration.

INTRODUCTION

Gasification and gas cleanup must be considered jointly, in view of today's clean environment regulations, because their combined action is required if clean gas is to be obtained from coal. Much of the coal sulfur appears in the gas; in addition, solid and tar particulates are present in concentrations that vary with the gasification process and coal composition. Gasification concepts undergoing development include gas cleanup in the overall processing scheme. Innovations are introduced mainly in the gasification step, and in some cases desulfurization is incorporated at this point. In other cases, gas purification could take place after gasification, and existing commercial systems may be satisfactory. However, new technology may be needed to meet more stringent demands.

Probably the least complicated system for converting coal into either low-Btu fuel gas or high-Btu pipeline gas is the one described herein. It bears the suggested name MORGAS (Morgantown gas). It incorporates pressure gasification in a stirred bed of mine-run coal, which may have any free-swelling index from low to high. Hydrogen sulfide is removed by contacting the hot gas with a bed of solid sorbent containing iron oxide; elemental sulfur is recovered during regeneration of the sorbent. These two basic elements can be combined with other unit operations as required by the end use of the gas.

EQUIPMENT

The Bureau's stirred-bed producer resembles the conventional fixed-bed producer except it uses mechanical deep-bed agitation. Principal dimensions and layout are shown in Figure 1. The fuel is supported on a revolving grate with an area of 9.6 ft^2. The bed depth varies between 6 and 7 feet; the depth is maintained by frequently adding coal in batches weighing 200 to 250 lb.; cinder is removed as needed. The water-cooled stirrer is balanced by a counter weight and supported in the pressure vessel by a thrust bushing sealed by a packing gland. Compound motion is imparted by combined horizontal rotation and vertical reciprocation, which can be controlled with respect to speed of rotation and vertical movement. Figure 2 shows the stirrer in greater detail. The two lower arms are water cooled, but the top arm is not cooled as it normally remains in a reduced temperature zone. Steady gasification conditions usually have been obtained by rotating the stirrer at one-half revolution per minute and limiting the vertical travel through a vertical distance of 2 feet. In practice, the stirrer passes through the bed in 15 minutes, but this rate can be slower or faster, as optimum rate varies with coal properties. The lowest point reached by the stirrer is usually set 2 feet above the top of the grate, but the limit of travel is within 1 foot of the grate.

Nuclear density gauges are used as shown in Figure 3 to indicate conditions within the pressure vessel. Ash zone, bed level, and voids in the bed are detected. Control of operating conditions are simplified by the use of these instruments; centralized, fully automated controls seem to be feasible for multiple units.

Continuous stirring of the bed maintained a dense fuel bed, giving good quality gas having constant composition. Vertical movement was as important as rotation for operating the experimental producer, but vertical movement may not be necessary for full-size units. Stirring was needed to break large clinkers that formed in the combustion zone, as well as coke in the gasification zone. As shown in Figure 4, the torque applied to rotate the stirrer varies directly with the bed depth covering the stirrer. At maximum depth the normal torque was 1,300 foot-pounds, reaching momentary peaks of 1,700 foot-pounds. Measurements were obtained on Upper Freeport coal, which gives a very hard coke. No significant difference in the torque load was found for double-screened 1/4- x 1-1/2-in. Upper Freeport and run-of-mine 0- x 1-1/2 in. Gasifying mine-run coal in a stirred bed was a significant advancement because the size limitation, heretofore believed necessary, can be eliminated. More of the market supply will be available for gasification, and preparation will be less costly. A screen analysis of run-of-mine Upper Freeport coal is given in Table 1. Twenty-five percent of the sample passed through a 1/16-in. sieve and 5 percent through 100 mesh. Some fine coal particles were entrained in the gas, but most were removed by a cyclone separator. Gas vented to the atmosphere and burned had a dust loading of about 0.5 to 0.7 lb/1000 ft^3.

Table 1. SCREEN ANALYSIS, UPPER FREEPORT COAL[a]

Screen size	Analysis, %	
	Direct	Cumulative
2-1/2 x 2 in.	2.5	2.5
2 x 1 in.	12.1	14.6
1 x 1/2 in.	12.2	26.8
1/2 x 1/4 in.	17.6	44.4
1/4 x 1/16 in.	30.2	74.6
1/16 in. x 50 mesh	16.9	91.5
50 x 100 mesh	3.5	95.0
100 x 200 mesh	2.1	97.1
200 mesh x 0 in.	2.9	100.0

[a] Free swelling index No. 8-1/2

RESULTS

Experimentally determined gas yields for moderately caking Illinois No. 6 coal are shown in Figure 5. Gas production was limited when the gas flow reached a velocity at which loss of fuel by entrainment becomes excessive.

This plot shows that the quantity of air limiting gas yield increases with increased pressure.

A mixture of iron oxide (hematite Fe_2O_3) and fly ash was the best sorbent found among more than twenty materials tested. Primary requirements were that the sorbent be readily available and relatively inexpensive, have reasonable sorption capacity and useful life, be easily regenerated for repeated use, and be resistent to fusion or disintegration over the useful temperature range. Fly ash as received could be formed into a durable and regenerably sorbent, but its sorption capacity was improved by adding iron oxide, increasing the concentration to 36 from 15 percent originally present. Other oxides present and inactive included silica 35 percent, alumina 18 percent, and small percentages of oxides of calcium, magnesium, sodium, potassium, and titanium. Iron oxide concentrations greater than 40 percent were unsatisfactory because the bed fusion temperature was lowered and fusion took place during normal operations.

Pilot quantities of the fly ash-iron oxide sorbent were made by two catalyst manufacturers by mulling and extruding the mixture to form 1/4-in. diameter cylinders with 1/4 to 1/2-in. lengths, which were then sintered to develop hardness. Mercury porosimeter measurements showed pore volume of new sorbent was 0.36 cm^3/g, but this decreased to 0.13 cm^3/g and remained constant after 30 regenerations, as shown in Figure 6. Surface area measured by nitrogen absorption ranged from 4.2 to 6.5 m^2/g. Sorption of hydrogen sulfide from dry simulated producer gas is given in Table 2 for materials of essentially the same composition but made by three laboratories.

Table 2. SORPTION OF H_2S FROM DRY PRODUCER GAS BY SINTERED IRON OXIDE-FLY ASH[a]

	Surface area,[b] m^2/gram	Bed temperature, °F	g S removed/100 g sorbent	
			From sorption data	From regeneration data
Commercial laboratory 1	6.5	1000	12.5	12.4
		1250	14.7	13.9
		1500	22.2	22.0
Commercial laboratory 2	4.2	1000	7.5	6.7
		1250	11.5	11.0
		1500	22.5	17.4
Mechanically formed by MERC[c]	4.2	1000	10.5	10.9
		1500	27.6	25.6

[a] All at 3 psig sorption pressure.

[b] BET nitrogen sorption method.

[c] Morgantown Energy Research Center, Morgantown, W. Va.

The sorbent made by MERC was tested through 174 regeneration cycles using simulated producer gas and bed temperatures of 1100, 1250, and 1500°F. Producer gas contains about 5 to 10 percent steam by volume, as excess steam is used to reduce temperature in the combustion zone, and the gas leaves the generator at a temperature around 1200°F. Steam amounting to 7 percent by volume was added to the gas for many of the above tests to closely simulate producer gas. Results obtained with gas containing steam, Figure 7, indicate a reduction in capacity when compared with capacities for dry gas as shown in Table 2. This was attributed to the lowering of the hydrogen sulfide concentration at the gas-solid interface by the added steam. Improving the mass transfer coefficient by raising the bed temperature was effective in increasing the capacity from 6 g sulfur/100 g sorbent at 1100°F to 10 g at 1500°F.

Iron oxide catalyzes the water gas shift reaction, $H_2O + CO = H_2 + CO_2$, and steam in producer gas affected the composition of the producer gas in passing through the sorption bed. The composition change resulting from the shift reaction was determined at 300 psig and temperatures of 1100, 1300, and 1400°F by passing producer gas containing 18 mole-percent steam through a bed of iron oxide fly ash sorbent using 1000 space velocity. Heating value was decreased by dilution from the carbon dioxide added to the gas; increased hydrogen and decreased carbon monoxide concentrations resulted in virtually no net change in heating value because they have nearly the same value, 319 and 316 Btu/scf, respectively. The shift would be beneficial if pipeline gas is the end use, because additional shifting would be needed to bring the hydrogen to carbon monoxide ratio close to 3:1. Increasing temperature favors higher carbon monoxide concentration at equilibrium. Results are shown in Table 3.

Table 3. CHANGE IN COMPOSITION AT 300 psig AND 1000 SPACE VELOCITY

	Hydrogen, %	Carbon Dioxide, %	Nitrogen, %	Carbon Monoxide, %	Heating Value, Btu/scf
Feed gas	17.0	6.7	50.9	25.4	134
Effluent gas:					
1100°F (dry)	16.4	7.9	51.0	24.7	130
1100°F (wet)[a]	22.9	13.2	51.1	12.8	113
1300°F (wet)[a]	21.3	11.0	51.0	16.7	121
1400°F (wet)[a]	19.9	9.7	51.0	19.4	125

[a] Steam content 18 volume-percent. Composition and heating value on dry basis.

Two sorption-regeneration cycles were completed, and cleaning gas was generated in the pressurized gas producer using Upper Freeport coal; the results are shown in Figure 8. Gas from the producer was transferred to the sorbent bed at system pressure of 120 psig via a heated pipeline. Bed temperatures were controlled to give 1100 and 1200°F, and flow rates were adjusted to give hourly space velocities of 710 and 940, respectively. Hydrogen sulfide concentration averaged 380 gr/100 ft^3; the gas contained approximately 0.516 dust, 1 lb tar, and 5 lb steam/1000 ft^3 Hydrogen sulfide in the gas leaving the sorbent bed had its concentration reduced to 10 and 20 gr/100 ft^3 and did not increase until after 6 hours on steam. Removal was 95 and 97 percent effective with respect to hydrogen sulfide. Tar was not removed by the sorbent.

Reaction mechanism is chemisorption, whereby hydrogen sulfide difusses throughout the sorbent and reacts with Fe_2O_3 forming FeS and FeS_2. Analyzing the spent sorbent indicated the empirical composition was $FeS_{1.3}$. Iron oxide, Fe_2O_3, was regenerated and the sulfur released as SO_2 by passing air or oxygen over the hot bed. With oxygen regeneration, the effluent gas was pure SO_2 until some oxygen passed through unreacted after regeneration was 90 percent complete. Rather than recovering the SO_2 as sulfuric acid or ammonium sulfate, it appears possible to reduce SO_2 to elemental sulfur. This may be done by regenerating two beds of saturated sorbent at the same time. Oxygen or air is supplied to one bed where the sulfur is oxidized,

$$4\,FeS + 7O_2 = 2\,Fe_2O_3 + 4\,SO_2, \qquad (1)$$

and the SO_2-rich effluent, free of oxygen, is supplied to the second bed where oxidation-reduction at 1500°F gives elemental sulfur,

$$3\,FeS + 2\,SO_2 = Fe_3O_4 + 5\,S. \qquad (2)$$

Before returning the second bed to sorption duty, magnetite is oxidized to hematite, as follows:

$$2\,Fe_3O_4 + 1/2\,O_2 = 3\,Fe_2O_3. \qquad (3)$$

CONCLUSION

Results indicate that hydrogen sulfide can be removed from producer gas by chemisorption using sintered pellets of iron oxide-fly ash. Long life is indicated for the sorbent used in a fixed bed. Fluidized- or expanded-bed operation may be possible if the pellets are reduced in size and shaped as spheres. The gas is generated and cleaned at pressure and temperature, thus conserving space and energy expended in gas compression. The initial results indicate that elemental sulfur may be recovered.

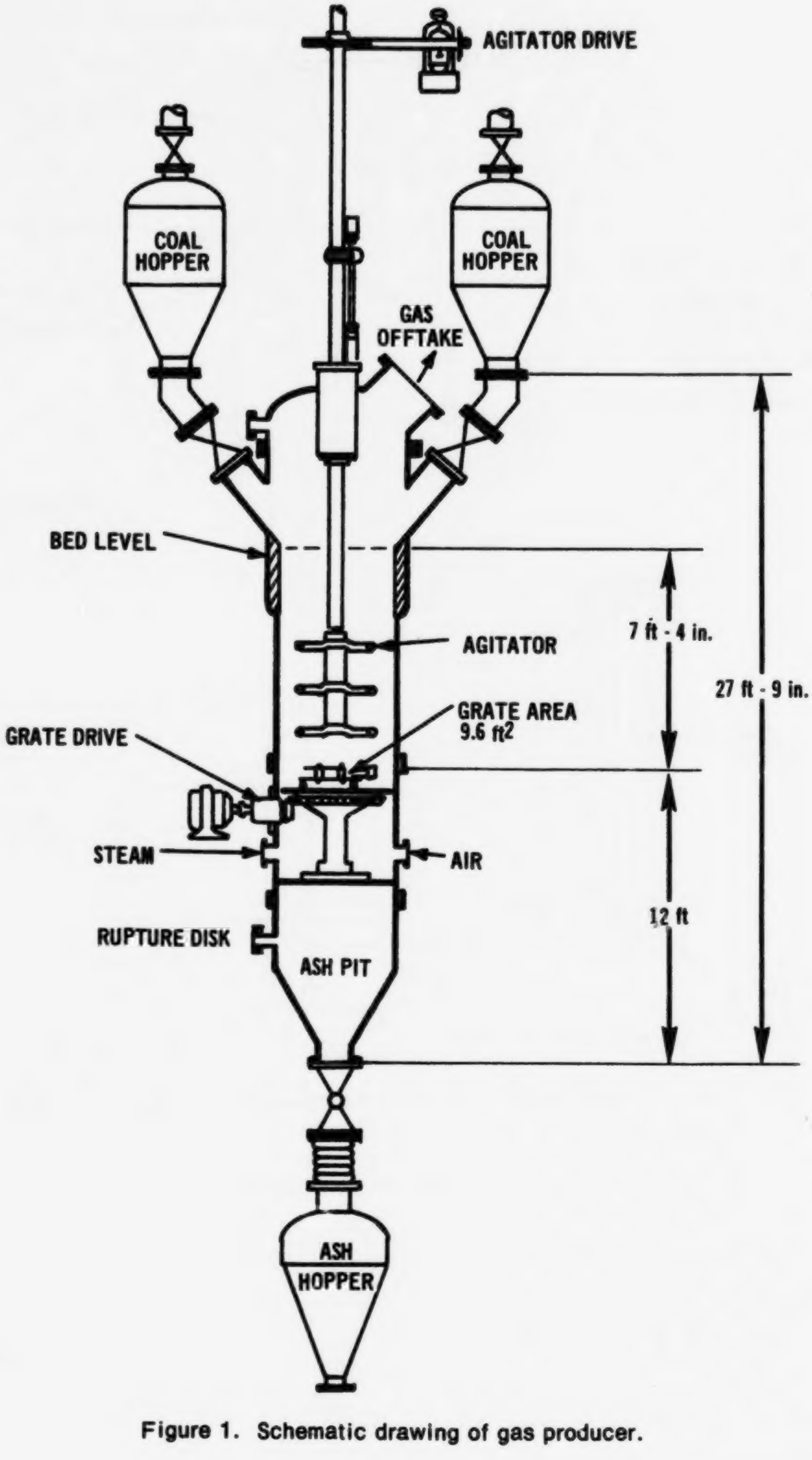

Figure 1. Schematic drawing of gas producer.

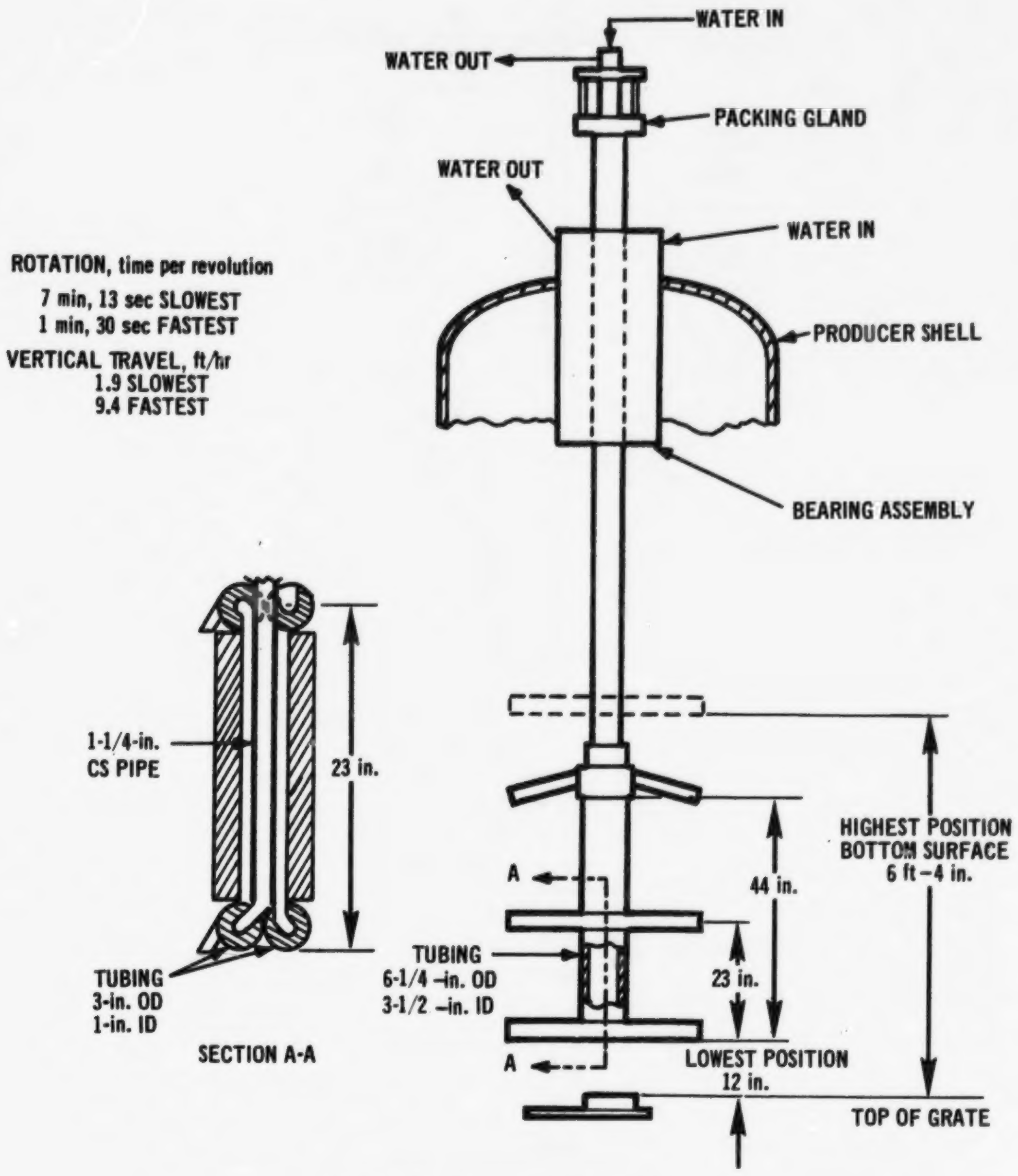

Figure 2. Stirrer arrangement.

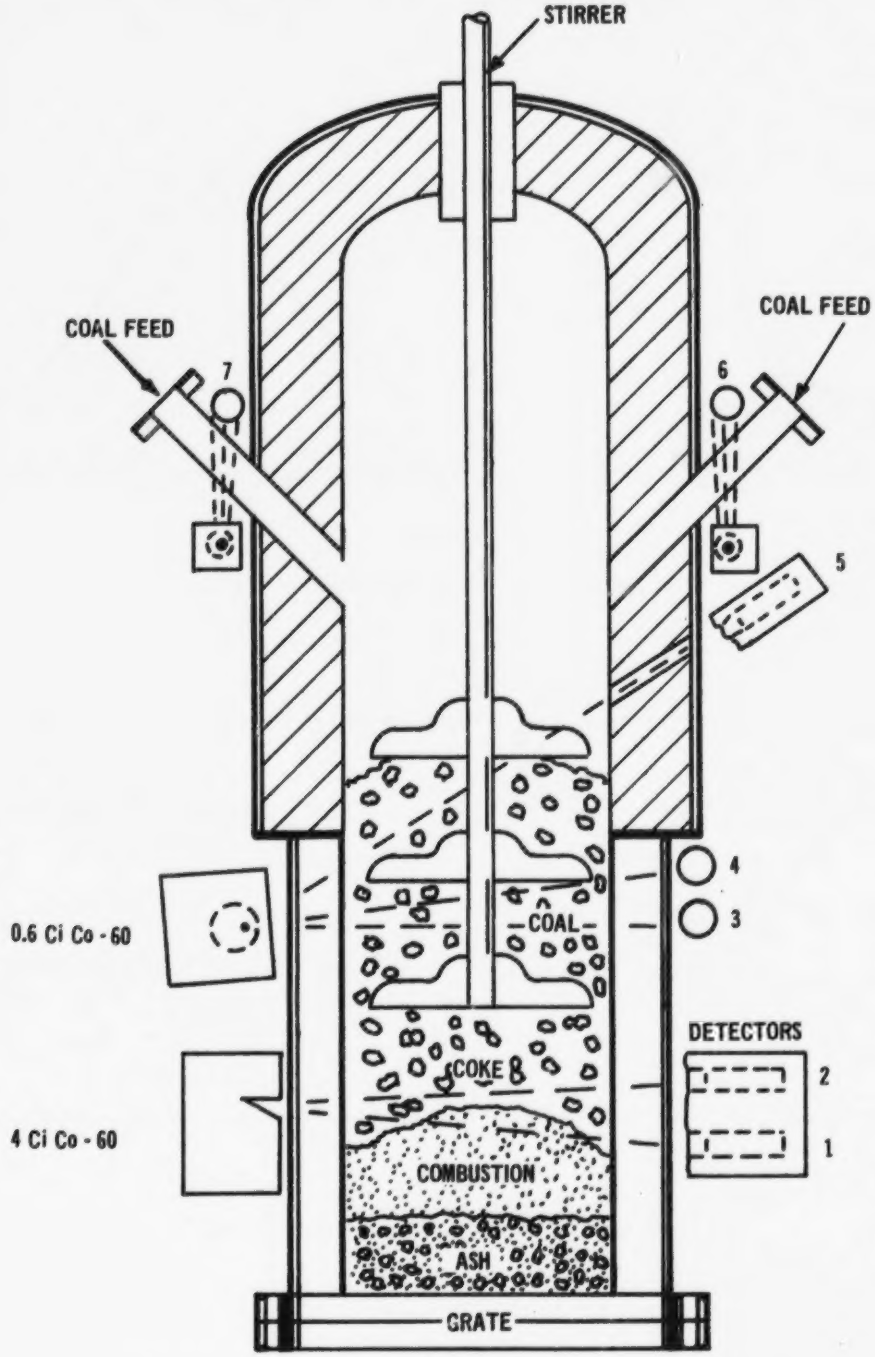

Figure 3. Nuclear density gauges applied to gas producer.

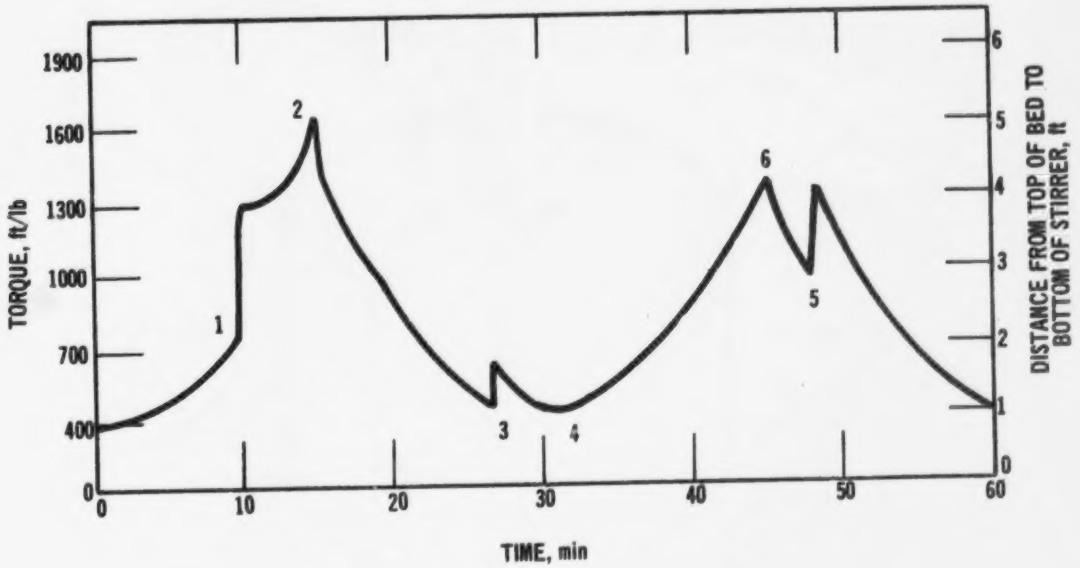

Figure 4. Torque applied to stirrer.

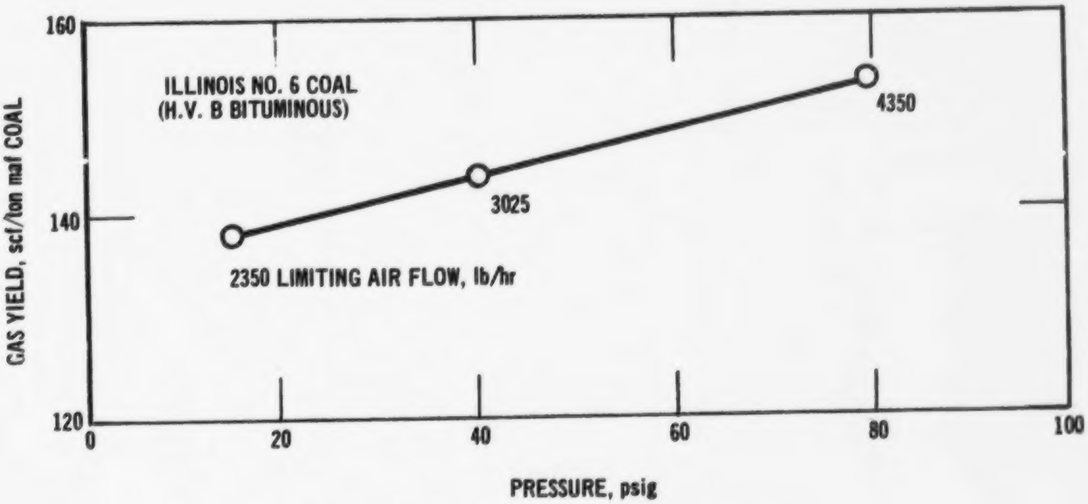

Figure 5. Pressure raises limiting air flow.

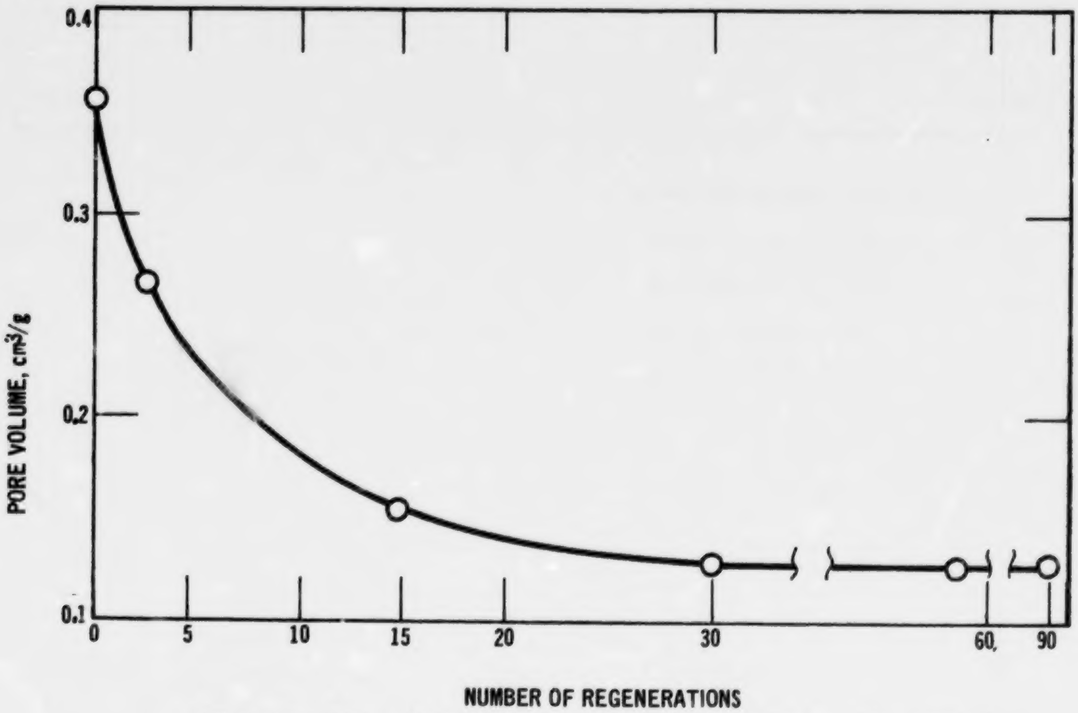

Figure 6. Pore volume reaches constant value after 30 regenerations.

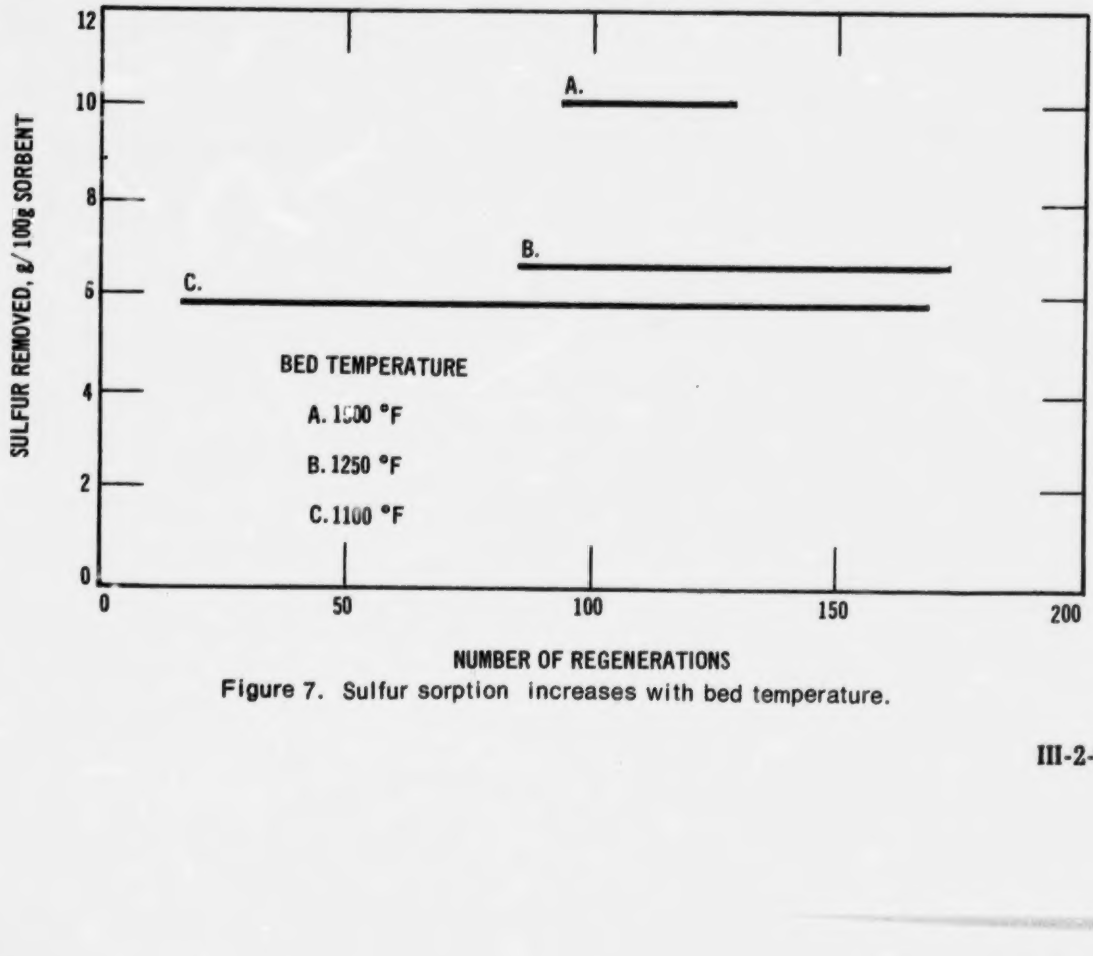

Figure 7. Sulfur sorption increases with bed temperature.

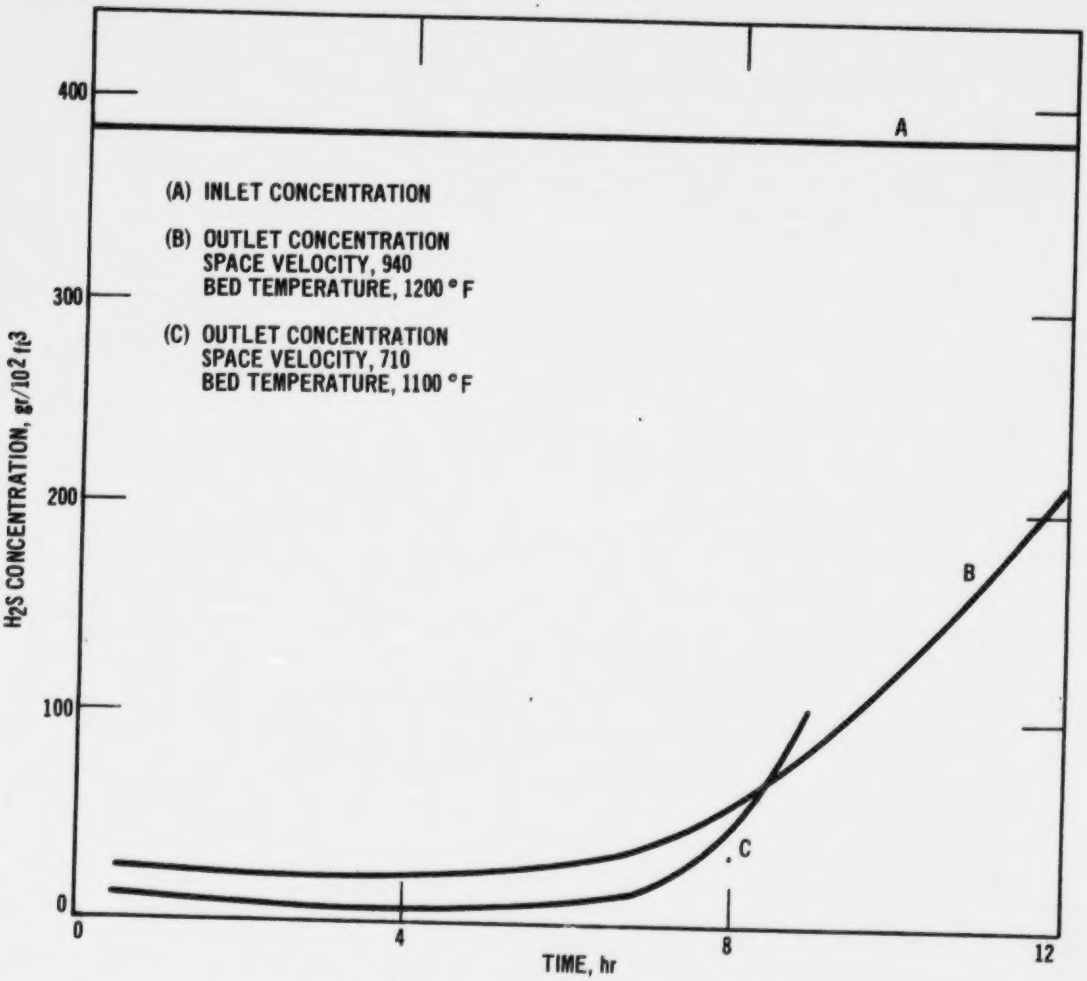

Figure 8. Removing H_2S from producer gas.

3. COAL GASIFICATION FOR CLEAN POWER GENERATION

D. H. ARCHER, E. J. VIDT, D. L. KEAIRNS, J. P. MORRIS AND J. L. CHEN

Westinghouse Research Laboratories

ABSTRACT

The growing demand for electrical energy in the U.S. requires the construction of new coal-fired power plants. Coal gasification, coupled with combined gas and steam turbine generation, provides a basis for a low cost, high efficiency, non-polluting plant. A fluidized-bed coal gasification process adapted to power generation has been devised. It uses air and steam for gasification and limestone or dolomite sorbent for desulfurization. A development effort is underway which includes the construction of a 1200 lb/hr coal gasifier and the performance of a supporting laboratory program.

INTRODUCTION

Need for Power Generation

In the next 20 years the quantity of electrical energy generated in the U.S. is expected to increase by a factor of almost 4, as shown in Figure 1.[1] Efforts to reduce the rate of growth in demand for electrical energy have been proposed. On the other hand it has been suggested that additional quantities of electrical energy will be required as programs are carried out to improve the environment and to maximize the efficiency of energy utilization. It seems prudent, therefore, to determine how the projected demands can best be met.

Both nuclear and fossil fuels—coal, oil, and gas—will be needed to supply this demand. Projections of fuel usage are made in Figure 2.[1] Nuclear fuel usage is limited by the number of nuclear power plants which can conceivably be constructed in coming decades. Natural gas shortages in the U.S. have led to the prediction that its use by utilities will be severely curtailed in the coming decade. Coal and oil, therefore, must provide the difference between total fuel demand for electrical generation and that portion supplied by nuclear fuel. An upper limit may be placed on imported oil to avoid problems resulting from dependence on foreign nations and from unbalance of payments in foreign trade. If so, the use of coal in power generation must increase by a factor of 3 in the next two decades. But if coal is not available in sufficient quantities to meet the demand, the use of oil in power generation will of necessity continue to increase.

Additional power plants must be constructed to meet the demands for electrical energy, as illustrated in Figure 3.[1] The generating capacity must increase from 325 GW in 1970 to 1400 GW in 1990. Fossil fuels will be used primarily in intermediate and peak load plants. Intermediate plants vary their output during each day to match the varying demand; their overall electrical energy output is about 40 to 60 percent of the maximum (if the plant operated continuously at rated capacity). Peaking plants operate only seasonally or in emergencies to match the varying demands; their energy output is 2 to 20 percent of the maximum.

Criteria for Power Plant Concepts

In order to compete successfully with a conventional coal burning steam power plant with a stack gas scrubber for SO^2 removal, an improved power generation system should have lower capital costs, higher operating efficiencies, and pollutant emissions which meet established requirements. Targets have, therefore, been established for the overall economics and performance of a new power plant concept:[2]

1. Capital costs for 250 to 600 MW plants operating in 1975, less than $330/kW
2. Overall operating efficiencies greater than 39 percent
3. Sulfur dioxide emission less than 1.2, NO_x emission less than 0.7, and particulate emission less than 0.1 lb/10^6 Btu heat of combustion of the fuel; thermal discharges prevented by the use of cooling towers.

Proposed Power Plant Concept

An efficient, economic power plant burning coal and providing low cost electrical energy for intermediate or base loads can be provided by coupling a coal gasification and gas cleaning system with a combined gas- and steam-turbine generation plant as shown in Figure 4. A two-stage fluidized-bed process gasifies coal using air and steam at temperatures of 1400 to 2100°F and pressures of 10 to 20 atm. The process desulfurizes the fuel gases at high temperature, 1400 to 1800°F, using a limestone or dolomite sorbent. The resulting CaS is treated for disposal; or the sorbent is regenerated for return to the process, and sulfur is recovered. Particles are removed from the hot fuel gases by cyclones, pebble bed filters, or porous ceramic filters. Most of the fuel gases, 80 to 90 percent, flow to gas turbine combustors where they burn with excess air to provide hot gases for expansion in a gas turbine. The remaining 10 to 20 percent of the fuel gases flow to a heat recovery boiler which provides steam at 1200 psig and 950°F to the steam turbine. About half the electrical energy output from the plant is produced by the gas turbine generator; and half by the steam turbine generator. The gas turbine also drives the main compressor for air flowing to both the combustor and the gasifier. A booster compressor for the air flowing to the gasifier is used to overcome pressure losses in the gasification and gas cleaning systems.

Similar combinations of coal gasification, gas cleaning, and combined gas and steam turbine generation have been proposed and explored.[3,4] These differ in the type of gasifier proposed, in the design of the gas cleaning system, and in the configuration of the power generation system. For example, STEAG[5] has built a plant employing fixed-bed coal gasifiers, low temperature aqueous scrubbers (for particulate removal — desulfurization is not included in the plant), pressurized boilers (for combustion of the fuel gases), and a gas turbine expander.

GASIFICATION FOR POWER PRODUCTION

General Background

Coal gasification for power production produces a clean fuel gas — with minimal sulfur and ash — which can be utilized either at atmospheric pressure in a conventional gas-fired boiler or at elevated pressure in a gas turbine combustor. Gas turbine generator or combined gas and steam turbine generator plants are preferred for new installations, because estimated costs for such plants are appreciably lower than those for conventional steam power plants. Gasification may also be useful in the future preparation of a clean fuel for an MHD[6] or fuel cell power plant.[7]

In gasification, air (or oxygen) is supplied to fuel in a quantity insufficient to complete the conversion of its carbon and hydrogen to CO_2 and H_2O. A number of possible sequential processes become important. Some of the oxygen added to the fuel reacts to form CO_2 and H_2O:

$$C + O_2 \rightarrow CO_2$$

$$H_2 + 1/2\,O_2 \rightarrow H_2O \qquad (1)$$

(oxidation)

These reactions release large quantities of heat. But unburned carbon from the fuel remains, and it reacts with CO_2 and H_2O to form CO and H_2:

$$C + \left(\begin{matrix}CO_2\\ H_2O\end{matrix}\right) \rightarrow CO + \left(\begin{matrix}CO\\ H_2\end{matrix}\right)$$

(gasification) (2)

These reactions absorb large quantities of heat. Hydrogen can also react with carbon from the fuel to form methane:

$$C + 2\,H_2 \rightarrow CH_4$$

(hydrogasification) (3)

This reaction is moderately exothermic. Finally the fuel, when heated, can also undergo

$$\text{Fuel} + \text{heat} \rightarrow C + CH_4 + HC$$

(devolatilization) (4)

where HC indicates higher hydrocarbons and tars. This reaction may also yield heat.

In a gasification process all of these processes can occur simultaneously throughout a reactor, or each reaction may be localized in a region of a reactor or in a separate vessel. Most gasification processes, however, are carried out so that the heat released by oxidation, hydrogasification, and devolatilization balances the heat required by gasification and the sensible heat of the overall reaction products. This overall heat balance can be achieved by controlling the amount of air (or oxygen), the amount of steam, or the amount of an inert gas added to the gasifier. If the reactions are carried out in separate regions or reactors, some mechanism is required to transfer heat between these regions.

The gas composition produced by a gasification process depends primarily on the nature of the fuel and on the temperature, pressure, and gas composition in the regions where gasification, hydrogasification, and devolatilization occur. These quantities determine the kinetic rates and the thermodynamic limits of the various processes — oxidation, gasification, hydrogasification, and devolatilization.

When H_2O, CO, H_2, and CO_2 coexist at high temperatures, they can also undergo the shift reaction:

$$H_2 + CO_2 \rightleftharpoons H_2O + CO. \qquad (5)$$

This reaction has a negligible heat effect, but its equilibrium does affect the gas composition according to the relative quantities of the gases involved in the shift.

In gasification, sulfur in the fuel is converted primarily to H_2S. A limestone/dolomite sorbent can be utilized in a fluidized bed to remove this pollutant from the fuel gases:

$$\begin{matrix}CaCO_3\\ CaO\end{matrix} + H_2S \rightarrow CaS + H_2O + \begin{matrix}CO_2\\ \text{—}\end{matrix} \quad (6$$

Over the past ten years much effort has been expended in the United States to develop processes to produce pipeline gas, consisting primarily of methane. Recently, interest in gasification processes to produce a fuel for power plants has increased. There are important distinctions between the gasified coal properties required for pipeline gas and those required for power plant fuel. These distinctions are both technical and economic; they have an important effect on the nature of the optimum gasification process for each of these applications.

In general, pipeline gas processes employ either pure O_2 or H_2 together, with H_2O at pressures well in excess of 20 atm to produce a product high in CH_4. Fuel gas for power processes uses air and H_2O at 20 atm or below to produce a lower-cost product. Table 1 summarizes the differences in the characteristic properties of a pipeline gas and a power plant fuel produced by a gasification process.

Table 1. FUEL GAS PROPERTIES REQUIRED FOR PIPELINE GAS AND FOR COMBINED CYCLE POWER PLANT FUEL

	Pipeline gas	Power plant fuel
Heat content, Btu/S ft^3	$\sim$1000	$>$ 90
Pressure, atm	$>$60	10-20
Temperature, °F	$\sim$70	70-1800[a]
Composition	Primarily CH_4	CO, H_2, N_2, CO_2, H_2O, CH_4
Cleanliness		
Sulfur	$<$1 ppm[b]	1.2 lb $SO_2/10^6$ Btu ($\sim$550 ppm)
Particulates	$<<$0.01 lb/10^6 Btu[b]	0.1 lb/10^6 Btu
Fuel cost target, \$/10^6 Btu	0.50-1.00	0.25-0.50

[a] A high temperature is advantageous and may be necessary if the heating value of the gas is low.

[b] Limits established by process requirements.

Fuel is currently processed in three reactor types — fixed bed, suspended bed, and fluidized bed. In a fixed-bed reactor, gases pass through a bed of solids at a velocity sufficiently low that the solid particles are not blown from the bed and are not supported by the flowing gases. The weight of the particles rests primarily on other particles which make up the bed. A boiler with a chain grate stoker is one type of fixed-bed reactor.

In a suspended-bed reactor, gases flow at a sufficiently high velocity that solid particles are carried along with the gases; their weight is supported by drag forces exerted by the gases. Contact between particles is limited to occasional collisions. A pulverized fuel boiler is one type of suspended-bed reactor.

In a fluidized-bed reactor, the gases flow through a bed of particles at a sufficiently high velocity to support their weight but not high enough to carry them out of the bed. Fluidized-bed gasification reactors have not yet been applied commercially to utility power generation. But at least five fluidized-bed gasification reactors are currently under development to produce pipeline gas and/or liquid fuels from coal. Other fluidized-bed gasification reactors are currently under development to produce pipeline gas from oil; fluidized-bed reactors are now used commercially in the catalytic cracking of oil, roasting of sulfide ores, incineration of oily wastes and sludges, production of organic chemical monomers, making of cement, conversion of nuclear materials for fuel elements, etc.

Fluidized-bed reactors provide the following features in processing solids and gases:

1. Ease and versatility in solids flowing and handling. Solid materials can readily be added to or removed from fluidized-beds. Gas velocities can be chosen to promote particles mixing in the bed or to cause separation between particles of different size and density.

2. Rapid heat transfer. The free movement of particles in a fluidized bed promotes rapid heat transfer both within the bed and between beds. Bed temperatures are therefore uniform and easy to control. Heat can be transferred between beds by the circulation of solids.

3. Effective gas-solid contact. Because the relative velocity between gas and solids is high, exchange of mass and heat is rapid. A fluidized bed also provides a large amount of solid surface in contact with flowing gas in a relatively small volume.

Table 2 summarizes various reactor types for coal gasification — their applications, advantages, and problem areas.

Table 2. REACTOR TYPES FOR COAL GASIFICATION

Reactor type	Application to gasification	Advantages	Problems
Fixed bed	Lurgi, McDowell Wellman gasifiers.	Developed technology; countercurrent flow of gas and solids possible.	Maintaining uniform gas and solids flow, adding coal, removing ash, temperature control, transfer of heat.
Suspended bed	Texaco partial oxidation, BCR two-stage gasifier.	High temperatures do not lead to excessive agglomeration of coal or ash.	Separating ash solids from gases, temperature control; co-current flow.
Fluidized bed	Consol gasoline and acceptor, IGT hydrogasification, FMC gasification, Bureau of Mines synthane gasifiers.	Versatility and ease of solids handling, uniform temperature; effective gas-solids contact.	Pretreatment to prevent coal agglomeration; multi-stage beds required to achieve counter-current flow.

The process of gasifying coal involves a number of process steps including:

Drying - The water content of the coal is reduced so that the particles are free-flowing and more readily transported and introduced into the gasification equipment.

Pretreatment - The coal is oxidized and/or devolatilized superficially in order to prevent sticking and agglomeration of particles.

Desulfurization - The sulfur released from the coal as H_2S during the gasification process is sorbed by limestone/dolomite particles of the bed.

Hydrogasification and devolatilization - Volatile products are driven off the coal in an atmosphere containing hydrogen, which reacts with the coal and char forming methane and higher hydrocarbons and releasing heat.

Gasification - Steam reacts with the char (or devolatilized coal) absorbing heat while forming fuel gases — H_2 and CO.

Combustion - Air reacts with the carbon of the char forming combustion products and releasing heat.

The rate and extent of each of these processes depends on temperature, pressure, atmospheric composition, and time. It appears advantageous and probably necessary to perform groups of these processes in individual reactors or in individual reaction steps within a single reactor; in this case provision must be made for the flow of reactants and heat between reactors or regions.

Proposed Coal Gasification Process for Electric Power Generation

A proposed improved multi-stage fluidized-bed coal gasification for power production process concept is illustrated in Figure 5. It comprises three process units — a dryer, a recirculating bed devolatilizer-desulfurizer, and a fluidized bed gasifier-combustor.

Crushed coal is dried in a fluidized bed and transported to the devolatilizer-desulfurizer unit. Here the devolatilization, desulfurization, and partial hydrogasification functions are combined in a single recirculating fluidized-bed reactor operating at 1300 to 1700°F. Dried coal is fed into a central draft tube of this reactor. In this tube, the coal feed and large quantities of recycled solids — char and/or lime sorbent — are carried upward by gases from the total gasifier flowing at velocities greater than 15 ft/sec. The recycle solids needed to dilute the coal feed and to temper the hot inlet gases flow downward in a downcomer — a fluidized bed surrounding the draft tube. These solids, flowing at rates up to 100 times the coal feed rate, effectively prevent or control agglomeration of the coal feed as it devolatilizes and passes through the plastic and sticky phase. A dense dry char is collected in the fluidized bed at the top of the draft tube. Lime sorbent is added to this bed in order to remove sulfur which is present as H_2S in the fuel gases. (An alternative concept would employ a separate desulfurization process. The fuel gases could be cleaned at high temperature in a fluidized bed of lime sorbent; or they could be cleaned, after cooling, at low temperature in a scrubber.) Spent (sulfided) sorbent is withdrawn from the reactor after stripping out the char either in the transfer line or in a separator of special design. Char is withdrawn from the top section of the bed. Heat is primarily supplied to this unit from the high temperature fuel gas produced in the total gasifier. Additional heat can be transported to the devolatilizer by solids carry-over in the gases from the total gasifier or by solids exchange between the two process units. Alternatively, additional heat can be generated in the devolatilizer by supplying air to the downcomers surrounding the draft tube.

The final gasification of the low sulfur char is conducted in a fluidized bed with a lower leg which serves as a combustor. In this section, char obtained from the devolatilizer-desulfurizer is burned with air at ~2100°F to provide the gasification heat. Heat is transported from the combustor to the gasifier both by combustion gases and by solids which flow up and down between the combustor and gasifier. The ash at this temperature agglomerates and segregates in the lower bed leg for removal. Gasification occurs in the upper section of the bed at 1800-2000°F with the sensible heats of both gas and char being used through solids exchange, to provide the heat requirements for the devolatilizer-desulfurizer.

This concept has the potential for overcoming the limitations of other gasification processes and providing a lower cost gasification system.

Utilization of wide variation in fuels — Caking coals and high ash coals can be used without costly and inefficient pretreatment. This feature is achieved by employing the recirculating bed to prevent agglomeration of coal particles.

Utilization of a wide variation in coal size - The sizing of the coal to the system is not critical. Coal with a size range of 1/8- to 1/4-inch x 0 can be used in the fluidized-bed system.

High thermal efficiency - Good heat economy is realized through the countercurrent movement of gases and solids between stages. The multi-stage arrangement provides the long residence time required for high carbon conversions, with good control over the temperature in both stages of gasification. Fluidized-bed gasification systems also provide a means for minimizing high carbon ash leaving the bed. This is achieved in the proposed design by using the agglomerating bed. Fluidized-bed agglomeration of coal ash with low carbon loss (<1 to 2 percent) has been demonstrated on both small and large scale equipment.

Reduced heat losses - Clean fuel gas can be produced without the heat loss as occasioned in cooling the fuel gases. The fluidized-bed concept permits fuel desulfurization by limestone or dolomite at elevated temperature.

Although this advanced gasifier concept is unique, it is composed of sub-systems which have been successfully operated by others. For instance, the recirculating bed has been developed by the Gas Council in England [8-13] and utilized by others.[14] The desulfurization step employing fluidized char and dolomite has been investigated by Consol Coal,[15] who embodied this idea predominantly for producing a low sulfur char; and by Esso (UK),[16] who uses fluidized beds of lime for gasifying and desulfurizing oil. Similarly, FMC employs multiple fluidized stages to produce char[22] Fuller, Chicago Bridge, Battelle,[19] and others [17,18,20,21] have used agglomerating fluidized-bed combustors in their processes. The use of multiple fluidized stages, with countercurrent flow of product gas, to achieve total gasification with desulfurization is a logical but novel method for utilizing all the inherent advantages of all these systems.

DEVELOPMENT PROGRAM

To realize this gasification concept and to achieve its potential benefits in power generation a development program is currently underway. It involves three parallel efforts:

1. The design, construction, and operation of a process development plant for gasifying 1200 lb coal/hr and for cleaning the resulting gases.

2. The planning and pursuit of laboratory studies in fluidization; H_2S sorption and lime regeneration; coal devolatilization, char gasification, and ash aggolmeration.

3. The conduct of systems studies on overall power plant performance and economics.

Process Development Plant Design

The purpose of the process development plant is to provide a means for investigating the gasification system — the devolatilizer-desulfurizer and the gasifier-combustor, both individually and in combination. The investigation is to:

1. Establish the operability of the equipment over a suitably wide range of conditions — flow rates, pressures, temperatures, and types of coal and lime sorbent;

2. Verify the suitability of the fuel gas produced for power production in a combined gas and steam turbine generator plant;

3. Produce the data required for engineering scale up and economic evaluation of the gasification equipment for a power plant.

Preliminary plans for the plant have been completed. These plans include flow diagrams (Figure 6) material and heat balances, and dimensional sketches for the devolatilizer-desulfurizer and the gasifier combustor. Special propane burners are included with the devolatilizer-desulfurizer to supply hot reducing gases so that this unit can be operated independently of the gasifier-combustor. If a supply of char is provided, the gasifier-combustor can also be operated independently of the devolatilizer-desulfurizer.

In addition to the devolatilizer-desulfurizer and the gasifier-combustor reactors, the process development plant includes:

1. Coal, char, and limestone sorbent receiving, storage, and feed systems;

2. Ash and spent sorbent removal, treating, and discharge systems;

3. Primary cyclones for removing particles from gases leaving the reactors; and

4. Gas scrubbing and quench systems for cleaning fuel gases prior to incineration.

Provisions have also been made for the addition of other features to the development plant including:

1. A secondary cyclone and filter to clean fuel gases leaving the primary cyclone of the devolatilizer-desulfurizer. These gases must be sufficiently clean to pass through a gas turbine with minimal corrosion, erosion, or deposition.

2. A gas turbine combustor to burn the clean fuel gases efficiently with a minimum production of NO.

3. A turbine blade test unit to demonstrate that the combustion gases have been effectively cleaned.

4. A lime sorbent regenerator to convert the spent (or sulfided) sorbent back to a form (a carbonate or oxide) which will absorb additional sulfur.

Operating conditions have been selected for the plant. Initially a coal feed rate of 300 lb/hr was selected as a basis for sizing the plant. Heat losses from the plant, however, were appreciable at this scale — amounting to 20 percent or more of the enthalpy exchanged between hot gases and coal in the devolatilizer-desulfurizer. Electrical heating was used as a means for minimizing this loss. Some of the critical internal dimensions of the reactors were also small — 2 inches or less. It was decided, therefore, to increase the capacity of the process development plant to 1200 lb coal/hr. Electrical heating is not required; minimum clearances of 3 to 4 inches are achieved. Finally, no increased cost of the plant is predicted. The reactor designs are simplified; outside dimensions of the pressure vessels remain unchanged. The solids feed and discharge systems are adequate for the increased capacity without modification.

The operating pressure for the gasification system is that required to supply fuel to the gas turbine combustor — a minimum of 10 to 16 atm. Current large industrial gas turbines use a combustor pressure of about 9 atm (gauge); advanced models in the next decade will probably use increased pressures, around 16 atm (gauge).

Operating temperatures have been estimated for the two reactors. The temperature for the devolatilizer has been chosen as 1600°F — high enough to crack higher hydrocarbons and thus to minimize carryover of tars.[23] This temperature also is close to that required to maximize the effectiveness of lime sorbents in sulfur removal.[16] A somewhat lower temperature — 1400°F — may still be effective for sulfur removal and would decrease the heat requirements for processing the coal in this reactor. The temperature for the combustor has been chosen as 2000-2100°F as required to agglomerate the coal ash.[19] The temperature in the gasifier will depend on the effectiveness of the solids in transferring heat between the combustor and the gasifier; the more effective the transfer, the smaller the temperature difference.

Gas velocities in the fluidized bed of the devolatilizer and gasifier (and thus bed diameters) have been selected on the basis of assumed particle size distributions and densities to achieve high capacities without excessive carry over of solids. Gas velocities in the draft tube and downcomer of the devolatilizer are chosen to achieve a ratio of about 80 to 1 of recycle solids to coal feed.[9-13] Finally, in the combustor gas velocities are chosen to minimize the quantity of char in the agglomerating section; it is expected that excessive char will inhibit agglomeration.

The depths of various bed sections have been chosen on the basis of various criteria. The combustor is deep enough to complete combustion of the char[24] and to capture 80 to 90 percent of the ash.[19] The gasifier is deep enough to react about half of the steam fed.[25,26] The devolatilizer is dimensioned to circulate the solids at the desired rate, to devolatilize the coal, and to remove 95 percent of the sulfur from the fuel gases.

The compositions of gas streams throughout the system have been estimated or selected on the basis of prior results or practice in coal gasification. Water gas equilibrium has been assumed to relate concentrations of H_2O, CO_2, H_2 and CO. It seems possible, however, that in this particular system greater amounts of the H_2O will react with the char due to the fluidization of this material[27] and greater quantities of CH_4 will be produced due to rapid heating of the coal to 1600°F in the presence of H_2.

Material and heat balances have been carried out which indicate that heat input to the devolatilizer-desulfurizer in addition to that provided by the hot gases from the gasifier may be required. This heat can be provided by exchange of solids between the gasifier and desulfurizer with additional heat generated directly within the devolatilizer by supplying air to the downcomer.

Detailed design of the process development plant is now about 50 percent complete. Figure 7 shows a model of the plant. Specifications for process vessels will shortly be ready to send to suppliers. The current schedule calls for mechanical completion of the plant in October 1973 assuming that complete funding is immediately available. The estimated cost of the plant is $4.2 million.

Laboratory Studies of Coal Gasification for Power Generation

To support the design of the process development plant and the evaluations of commercial performance and economics, laboratory studies are now underway in two areas:

1. Cold model studies of fluidized beds to study bed circulation, jet penetration, solids separation and elutriation, and solids attrition.
2. Limestone sorbent behavior studies to study sulfur capture, sorbent regeneration, and sorbent disposal.

Work will be initiated shortly in a third area:

Coal behavior studies to study devolatilization, char gasification, and ash agglomeration.

Cold model studies have been carried out in a two dimensional fluidized bed (Figure 8) to study the transport of particles upward in a draft tube and downward in a downcomer. The penetration of the jet of particles emerging from a draft tube into the fluidized bed at its upper end has also been studied. The transfer of particles from the downcomer to the draft tube at the base of the devolatilizer is now being observed. Correlations relating gas flow rates, particle flow rates, pressure drops, and bed dimensions have been produced. These correlations are useful in designing the devolatilizer and in determining recirculation rates within it from process measurements. Observations are now being carried out on bed slugging, solids separation and elutriation, solids attrition, and solids movement in fluidized beds simulating the gasifier.

Limestone and dolomite behavior has been studied in a thermogravimetric analyzer[29] (Figures 9 and 10), which measures weight changes of small samples of the solid as it is exposed to various atmospheres. The sorption of H_2S and the regeneration of the resulting CaS by H_2O and CO_2 have been studied at pressures, temperatures, and gas compositions projected for the process development plant. Both the degree of sorbent utilization and the rate of sorption and regeneration appear sufficiently high for the process to be practical (Figures 11 and 12). The oxidation of the sulfided CaS sorbent in air to $CaSO_4$ has also been studied as a means for rendering spent sorbent inert for disposal. A problem has been encountered in completing this conversion (Figure 13). Tests of sorbent behavior are continuing to determine optimum operating conditions for sulfur removal and regeneration, to estimate the number of sorption-regeneration cycles the sorbent can effectively undergo, and to develop an improved sorbent disposal process.

Systems and Economic Studies

Preliminary studies have been carried out to compare the cost of the gasification process proposed here with alternative fixed, suspended, and fluidized bed processes. It appears that cost reductions of 20 to 40 percent can be achieved by the development of the process. Cost calculations have also been made for overall plants comparing a coal gasification, combined gas and steam turbine cycle plant with a conventional steam plant using a stack gas scrubber for sulfur removal. An electric power plant using the coal gasification process is estimated to cost 20 to 30 percent less than the conventional plant.

CONCLUSION

A comprehensive program is underway to develop a coal gasification system for a combined gas and steam turbine generator plant. Such a power plant is expected to be lower in capital costs, lower in pollutant emissions, but equal in overall efficiency to a conventional power plant. The goal of the overall program is to complete the demonstration of such a plant before the end of this decade. Further improvements in gas turbine technology are expected which will improve the efficiency and reduce the costs for commercial power plants in the 1980's.

BIBLIOGRAPHY

1. Hauser, L.G., W.H. Comtois, and R.R. Boyle. The Effect of Fuel Availability on Future R & D Programs in Power Generation. (Presented at the American Power Conference. Chicago. April 18-20, 1972.)

2. Evaluation of the Fluidized-Bed Combustion Process. Summary Report, Vol. 1. Westinghouse Research Laboratories. Pittsburgh, Pa. Prepared for Office of Air Programs, Environmental Protection Agency, Research Triangle Park, N.C. under Contract Number CPA 70-9. October 1972.

3. Rudolph, P.F.H. New Fossil-Fueled Power Plant Process Based on Lurgi Pressure Gasification of Coal. (Presented at Joint CIC-ACS Conference, American Chemical Society, Toronto. May 24-29, 1970. pp. 13-38.)

4. Robson, F.L., A.J. Giramonti, G.P. Lewis, and G. Gruber. Technological and Economic Feasibility of Advanced Power Cycles and Methods of Producing Nonpolluting Fuels for Utility Power Stations. Final Report, United Aircraft Research Laboratories. East Hartford, Conn. Prepared for the National Air Pollution Control Administration, Durham, N.C. under Contract Number CPA 22-69-114. December 1970.

5. Bund, K., K.A. Henney, and K.H. Krieb. Combined Gas/Steam-Turbine Generating Plant with Bituminous-Coal High-Pressure Gasification Plant in the Kellermann Power Station at Lunen. (Presented at 8th World Energy Conference. Bucharest. June 28-July 2, 1971.)

6. Way, S. New Directions in Power Generation-MHD. In: Proceedings of North American Fuel Technology Conference, Ottawa, Canada, June 1970.

7. Keairns, D.L. and D.H. Archer. New Directions in Power Generation — Fuel Cells. In: Proceedings of North American Fuel Technology Conference, Ottawa, Canada, June 1970.

8. Dent, F.J. Methane from Coal. BCURA Quarterly Gazette, 42:1-14, 1960.

9. Dent, F.J., R.F. Edge, D. Hebden, F.C. Wood, and T.A. Yarwood. Experiments on the Hydrogenation of Oils to Gaseous Hydrocarbons. Midlands Research Stations, The Gas Council, Research Communication GC37, Inst. of Gas Engineers Trans., pp. 594-643.

10. Murthy, P.S. and R.F. Edge. The Hydrogenation of Oils to Gaseous Hydrocarbons. Midlands Research Station, The Gas Council, Gas Council Research Communication GC88, IGE Journal, August 1963, pp. 459-476.

11. Thompson, B.H., B.B. Majumdar, and H.L. Conway. The Hydrogenation of Oils to Gaseous Hydrocarbons. Midlands Research Station, The Gas Council Gas Council Research Communication GC122. IGE Journal. pp. 415-428, June 1966.

12. Horsler, A.G. and B.H. Thompson. Fluidization in the Development of Gas Making Processes. Midlands Research Station. The Gas Council pp. 51-59, March 1967.

13. Horsler, A.G., J.A. Lacey, and B.H. Thompson. High Pressure Fluidized Beds. Chemical Engineering Progress. 65(10):59-64, 1969.

14. Pilot-Scale Development of the CSF Process. R & D Reports Number 39, Volume IV, Book 3. July 1, 1968 - December 31, 1970. Consolidation Coal Company, Research Division, Library, Pa. Prepared for Office of Coal Research, Department of the Interior, Washington, D.C. under Contract Number 14-01-0001-310 (1).

15. Theodore, F.W. Low Sulfur Boiler Fuel Using the Consol CO_2 Acceptor Process. Report Number 2. Consol Coal. Prepared for Office of Coal Research, U.S. Department of the Interior, Washington, D.C. under Contract Number 14-01-0001-415, November 1967.

16. Craig, J.W., G.L. Johnes, G. Moss, J.H. Taylor, and D.E. Tisdall. Study of Chemically Active Fluid Bed Gasifier for Reduction of Sulphur Emissions. Final Report. Esso Research Center, Abingdon, Berkshire, England. Prepared for Office of Air Programs, Environmental Protection Agency, Research Triangle Park, N.C. under Contract Number 70-46. June 22, 1970 to March 1972.

17. Godel, A.A. Ten Years of Experience in the Technique of Burning Coal in a Fluidized Bed. Revue Generale de Thermique. 5:348-358, 1966.

18. Godel, A.A. and P. Cosar. The Scale-Up of a Fluidized Bed Combustion System to Utility Boilers. American Institute of Chemical Engineers Symposium Series. 67(116):210-218, 1971.

19. Goldberger, M.W. Collection of Fly Ash in a Self-Agglomerating Fluidized-Bed Coal Burner. (Presented at Winter Annual American Society of Mechanical Engineers Meeting. Pittsburgh. ASME paper 67-WA/FU-3. November 1967.)

20. Jequier, L., L. Longchambon, and G. Van de Putte. The Gasification of Coal Fines. J. Inst. Fuel. 33:584-591, 1960.

21. Jequier, L. et al. Apparatus for Dense-Phase Fluidization. U.S. Patent 2,906, 608, September 29, 1959.

22. Jones, J.F., F.H. Schoemann, J.A. Hamshar, and R.T. Eddinger. Char Oil Energy Development. Chemical Research and Development Center, FMC Corporation. Prepared for Office of Coal Research, U.S. Department of the Interior, Washington, D.C. under Contract Number 14-01-0001-498, October 1966 - June 1971.

23. Consol Coal, Private communication from E. Gorin and G. Curran.

24. Hoy, H.R. and A.G. Roberts. Fluidized Combustion of Coal at High Pressure. (Presented at Annual AIChE Meeting, San Francisco. November 1971.)

25. von Fredersdorff, C.G. The Reaction Rates Between Carbon-Carbon Dioxide and Carbon-Steam at 2000°F and 1 atm. I.G.T. Research Bulletin No. 19, 1955.

26. Blackwood, J.D. and F. McGrory. The Reaction Rates Between the Gas and the Carbon at 1600°F and High Pressure (1 to 50 atm). Australian J. Chemistry. 11:16-33, 1958.

27. Blackwood, J.D. and A.J. Ingerme. The Reaction Rates Between the Gas and the

Carbon at 1600°F and High Pressure (1 to 50 atm). Australian J. Chemistry. 13:194-209, 1960.

28. Squire, A.M., M.J. Gluckman, R.A. Graff, R. Shinnar, and J. Yerushalmi. Studies Toward Improved Techniques for Gasifying Coal. Part II: Technical Presentation. Submitted to the National Science Foundation (RANN) by The City College and Research Foundation of The City University of New York, June 1972.

29. O'Neill, E.P., D.L. Keairns, and W.F. Kittle. Kinetic Studies Related to the Use of Limestone and Dolomite as Sulfur Removal Agents in Fuel Processing. (Presented at 3rd International Conference on Fluidized Bed Combustion. Hueston Woods. October 29 - November 1, 1972.) (Session I, Paper 6, this document.)

APPENDIX A

Heat and Material Balances for 1200 lb/hr Process Development Plant

The heat and material balances pertain to a mode of operation in which air is fed to the downcomer of the devolatilizer-desulfurizer to generate part of the heat requirements of the vessel. Exchange of hot solids between the two reactors is another way to increase the heat input to the devolatilizer-desulfurizer; this mode is not compatible with the planned independent operation of the reactors, however.

Air fed to the downcomer reacts mainly with carbon since fuel gas is kept out of the zone by the flow of fluidizing gas. The air for combustion is introduced as part of the fluidizing gas. Rapid, countercurrent flow of solids and gas in the downcomer prevents excessive temperature rise.

The material balance is given in Table A1 and the individual flow streams are identified in Figure A1. Separate heat balances for the two reactors are presented in Table A2 and A3.

The heating value of the fuel gas product is dependent on the assumptions made and, in the present example, is 123 Btu/scf. Assumptions regarding stream temperatures, fines elutriation rates, and transport and fluidizing gas requirements are incorporated in Table A1.

Other assumptions follow:

1. Ultimate analysis of coal:

C	74.0 wt %
H	5.0 wt %
N	1.5 wt %
S	3.5 wt %
O	6.0 wt %
Ash	10.0 wt %
	100.0 wt %

2. The products of devolatilization, including decomposition of tar and oil, were estimated as follows:

The volatile matter includes all of the oxygen and hydrogen and half of the sulfur in the coal.

Oxygen divides equally between carbon and hydrogen in the volatile matter, forming CO and H_2O.

Sulfur is evolved as H_2S.

The remaining hydrogen forms H_2 and CH_4 in the molecular ratio of 2 to 1.*

Carbon in the volatile matter in excess of that producing CO and CH_4 reverts to solid carbon.

3. The heat of carbonization of the coal is small and can be neglected.

4. Twenty-three percent of the total char carbon derived from the coal is burned in the

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*Bituminous Coal Research, Inc., Gas Generator Research and Development. Survey and Evaluation, Phase One, Vol. Two. BCR Rept. L-156, pp. 223-5 (1965).

Table A1. MATERIAL BALANCE FOR 1200 lb/hr PROCESS DEVELOPMENT PLANT

Stream no.	1	2	3	4	5	6	7	8	9	10	11	12
Temperature, °F	77	1000	1600	1600	1000	1600	2000	1000	1000	1000	400	665
Solids	Coal	Calcined dolomite	Char	Fines	Ash	Spent dolomite	Fines	Char	Fines	—	—	—
Lb/hr	1200	696	440	560	120	659	280	440	560			
Composition, wt %												
Fixed carbon	55.0		82.3	66.7			50.0	83.3	66.7			
Volatile matter	35.0											
Ash	10.0		16.7	33.3	100		50.0	16.7	33.3			
MgO·CaCO$_3$		100				77.7						
MgO·CaS						22.3						
Gas	Transport	Transport	—	Fuel gas	—	Stripping	Gasifier product	Transport	Transport	Air	Steam	Fluid-izing
Lb/hr	480	47		7354		94	4680	88	112	3113	741	1546
Composition, mole %												
N_2	53.7	53.7		50.0		53.7	48.6	53.7	53.7			10.5
CO	19.0	19.0		17.7		19.0	19.5	19.0	19.0			
CO_2	9.2	9.2		8.6		9.2	8.8	9.2	9.2			
H_2	14.3	14.3		13.3		14.3	11.4	14.3	14.3			
H_2O	1.0	1.0		7.9		1.0	11.2	1.0	1.0			
CH_4	2.8	2.8		2.6		2.8	0.1	2.8	2.8			
H_2S							0.4					
Air										100	100	89.5

Table A2. COMBUSTOR-GASIFIER HEAT BALANCE (Btu/hr)

Input	
Air and steam preheat	827,400
Transport gas preheat	55,900
In char and fines feed	369,600
Reaction heat: $S \rightarrow H_2S$	5,700
$C \rightarrow CO$	1,633,300
$C \rightarrow CO_2$	2,649,500
$H_2O \rightarrow H_2$	-2,138,000
	3,403,400
Utilization	
Heat losses	321,900
Out in ash	25,800
Out in fines	167,100
Out in product gas	2,888,600
	3,403,400

Table A3. DEVOLATILIZER-DESULFURIZER HEAT BALANCE (Btu/hr)

Input	
In combustor/gasifier product gas	2,888,600
In fines from combustor/gasifier	167,100
In dolomite feed	165,600
In transport gas	38,400
In fluidizing gas	127,100
Combustion of carbon in downcomer	1,337,700
Water gas shift reaction	16,700
	4,741,200
Utilization	
Heat losses	267,000
Out in char and fines	493,700
Out in spent dolomite	298,300
Out in product gas	3,626,300
Desulfurization reactions	55,900
	4,741,200

downcomer and produces CO and CO_2 in the molecular ratio of 2 to 1.

5. Recycled fuel gas, after drying to 1 percent moisture, is used to transport solids and to strip char from the spent dolomite.

6. The concentration of CO_2, CO, H_2O, and H_2 in the product streams of the two reactors are related in accordance with the water gas shift equilibrium.

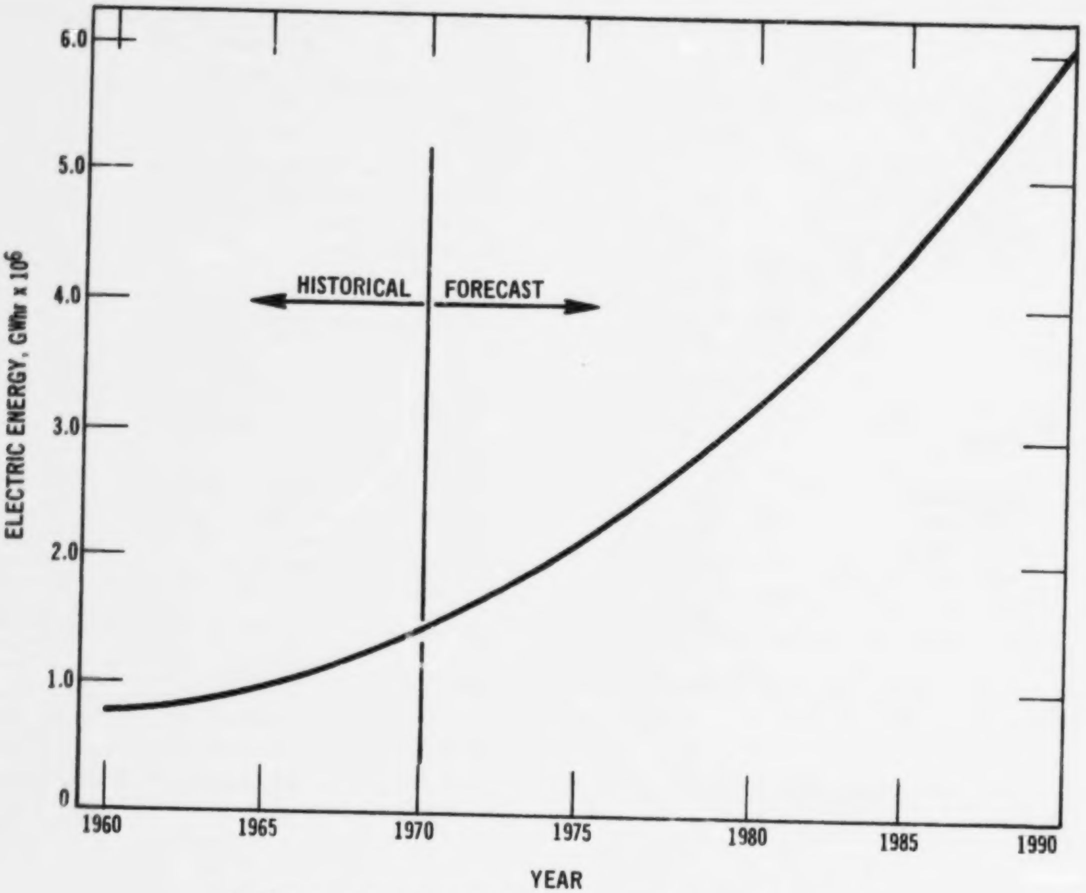

Figure 1. Annual electric energy generation in the U.S.

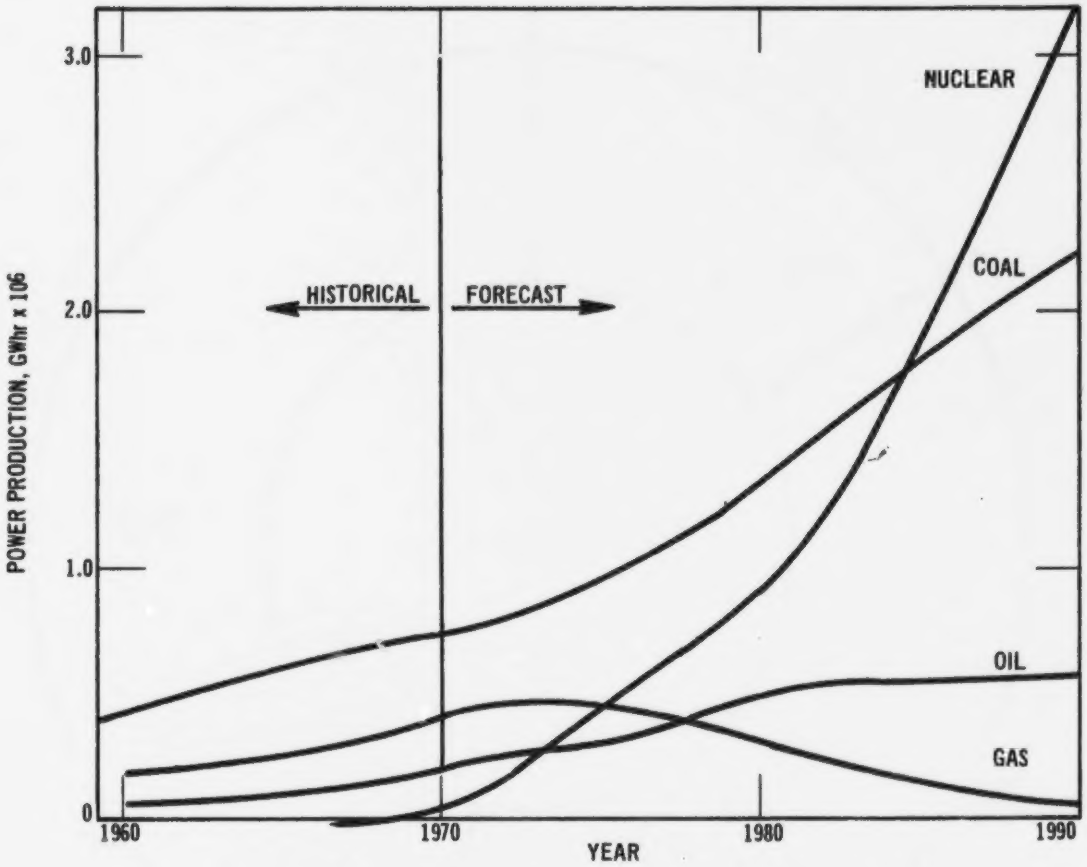

Figure 2. Forecast of power generation by fuel in the U.S.

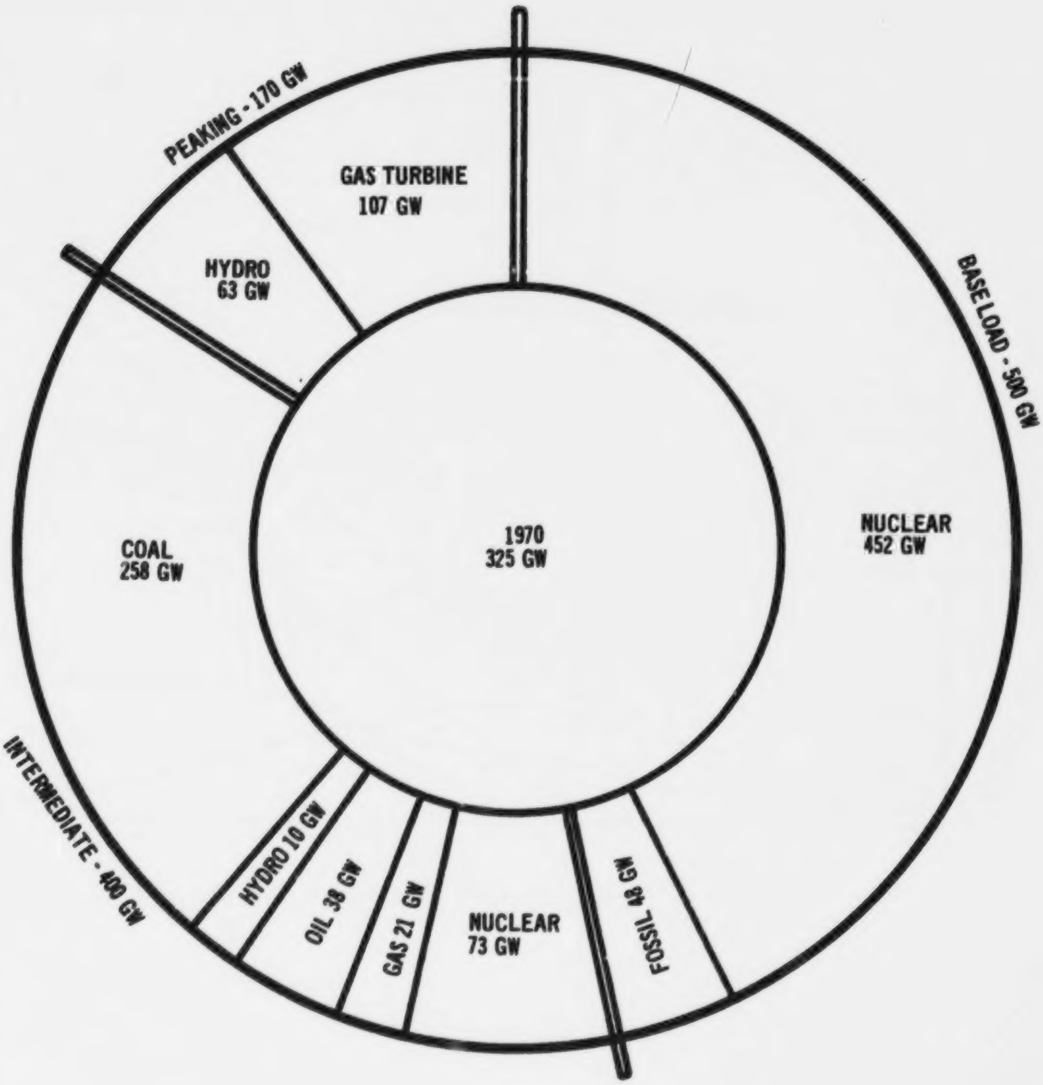

Figure 3. Generation additions by fuel and type in the U.S., 1970-1990.

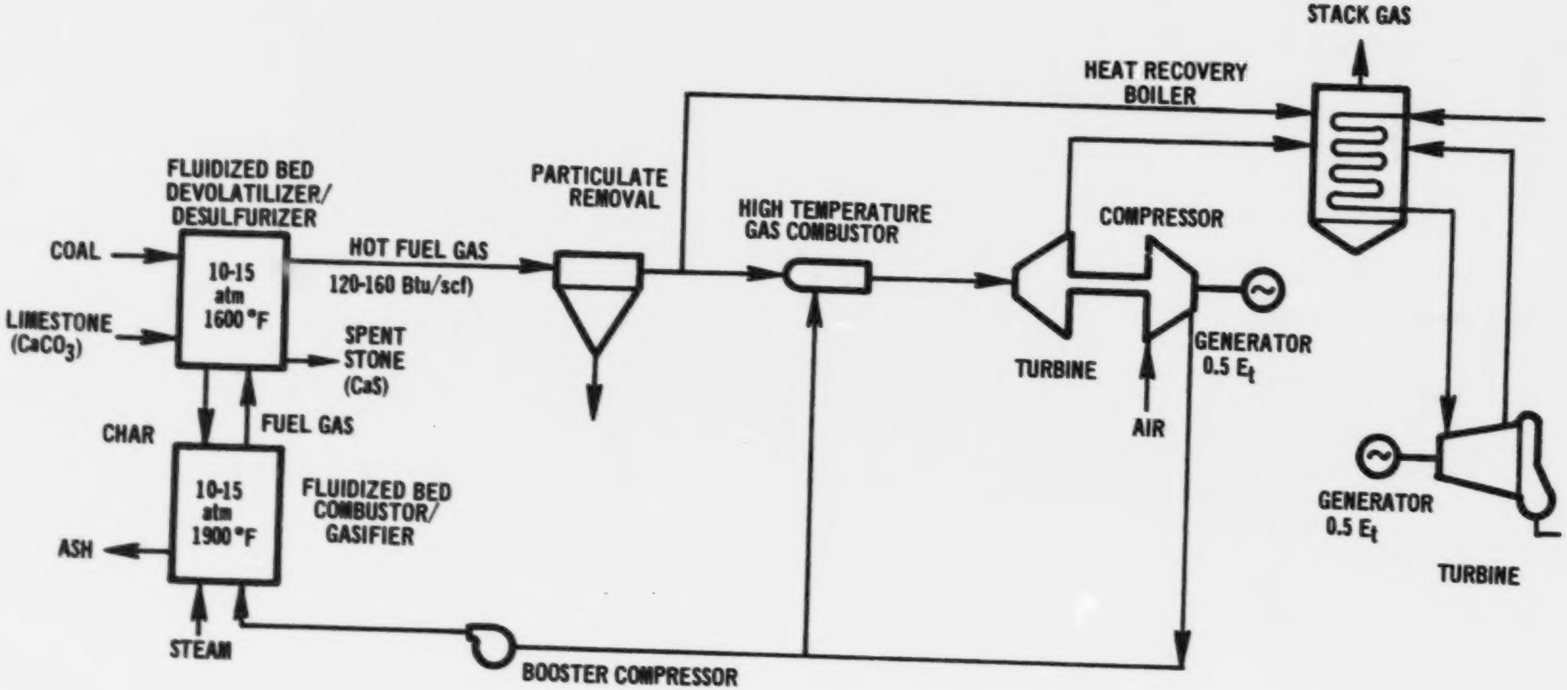

Figure 4. Coal gasification - combined cycle plant.

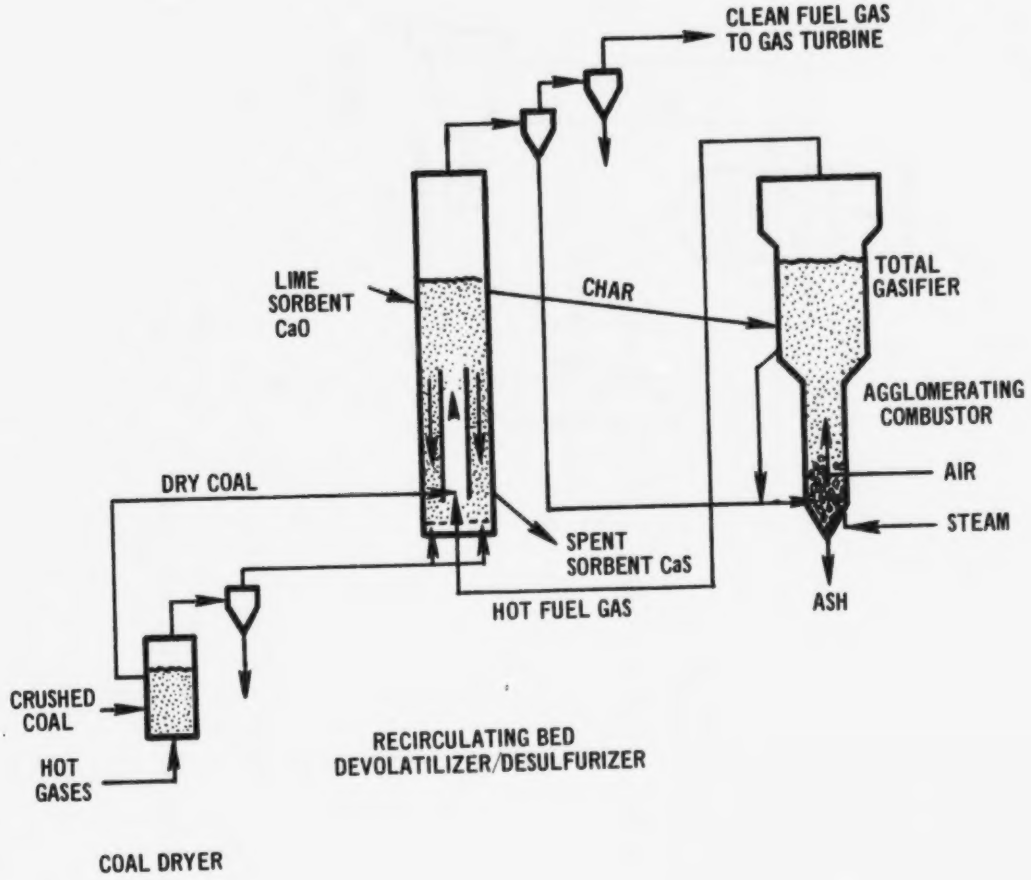

Figure 5. Westinghouse multistage fluidized-bed gasification process.

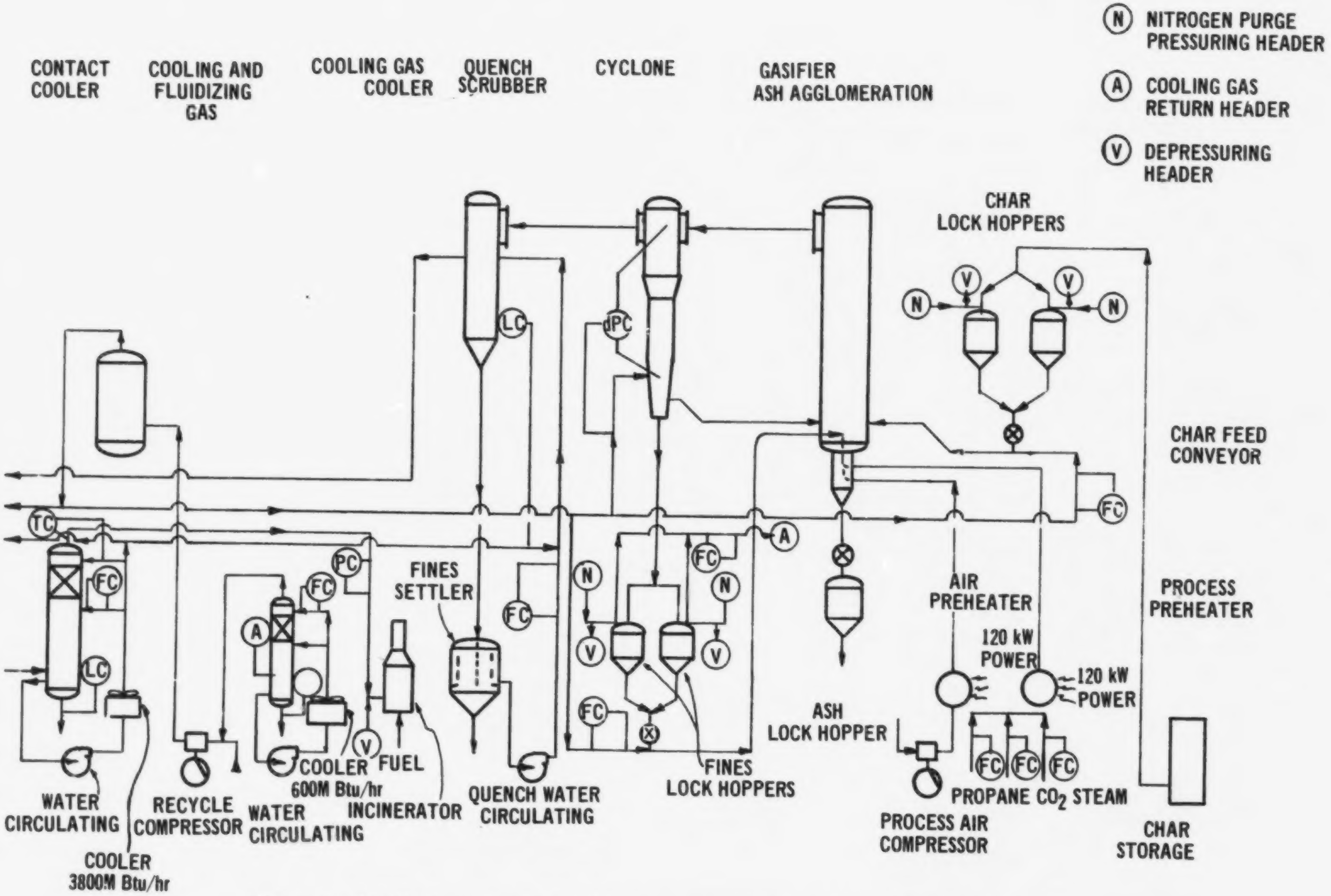

Figure 6. Westinghouse coal gasification process development plant flow sheet.

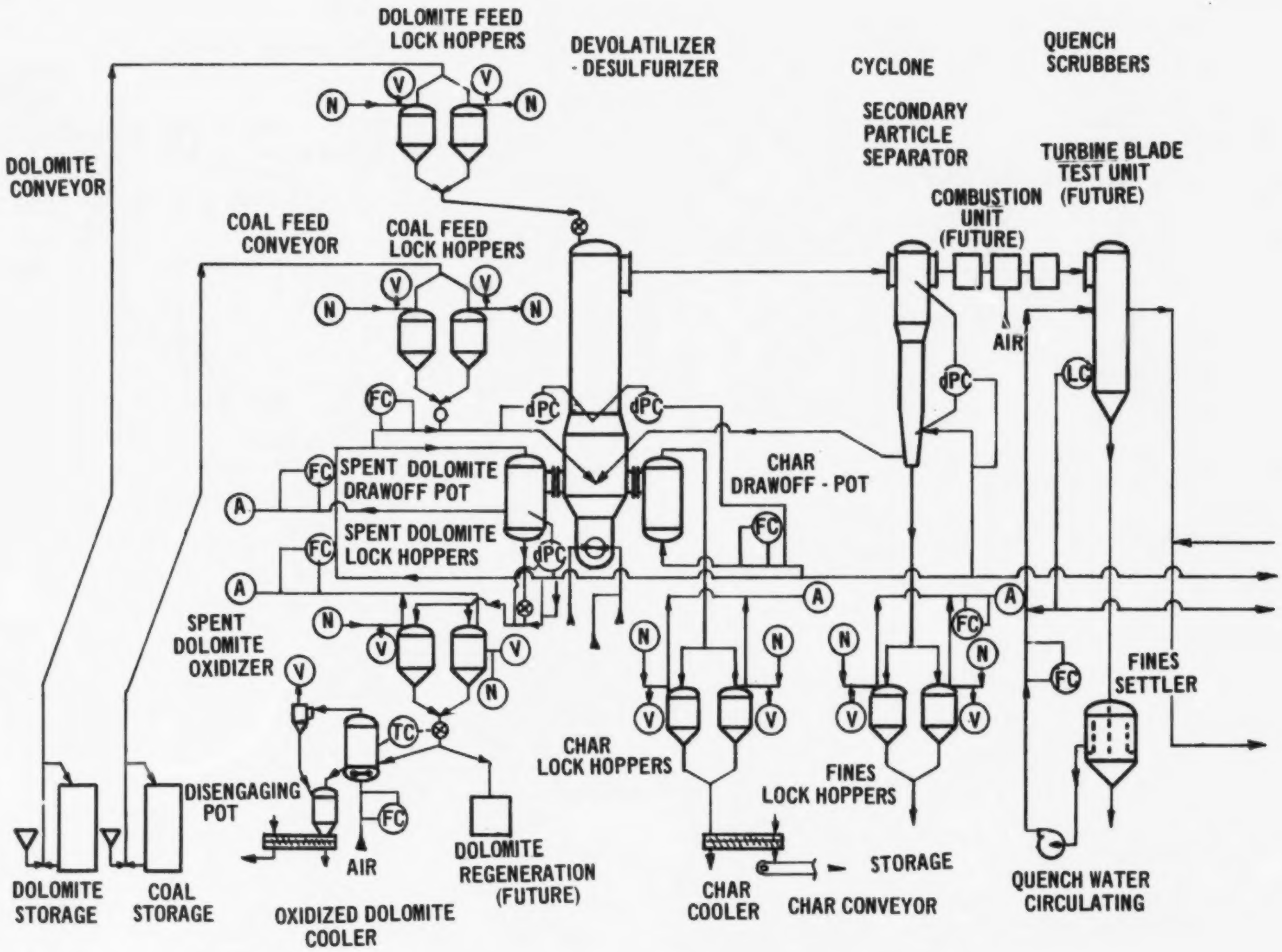

Figure 6. Westinghouse coal gasification process development plant flow sheet.

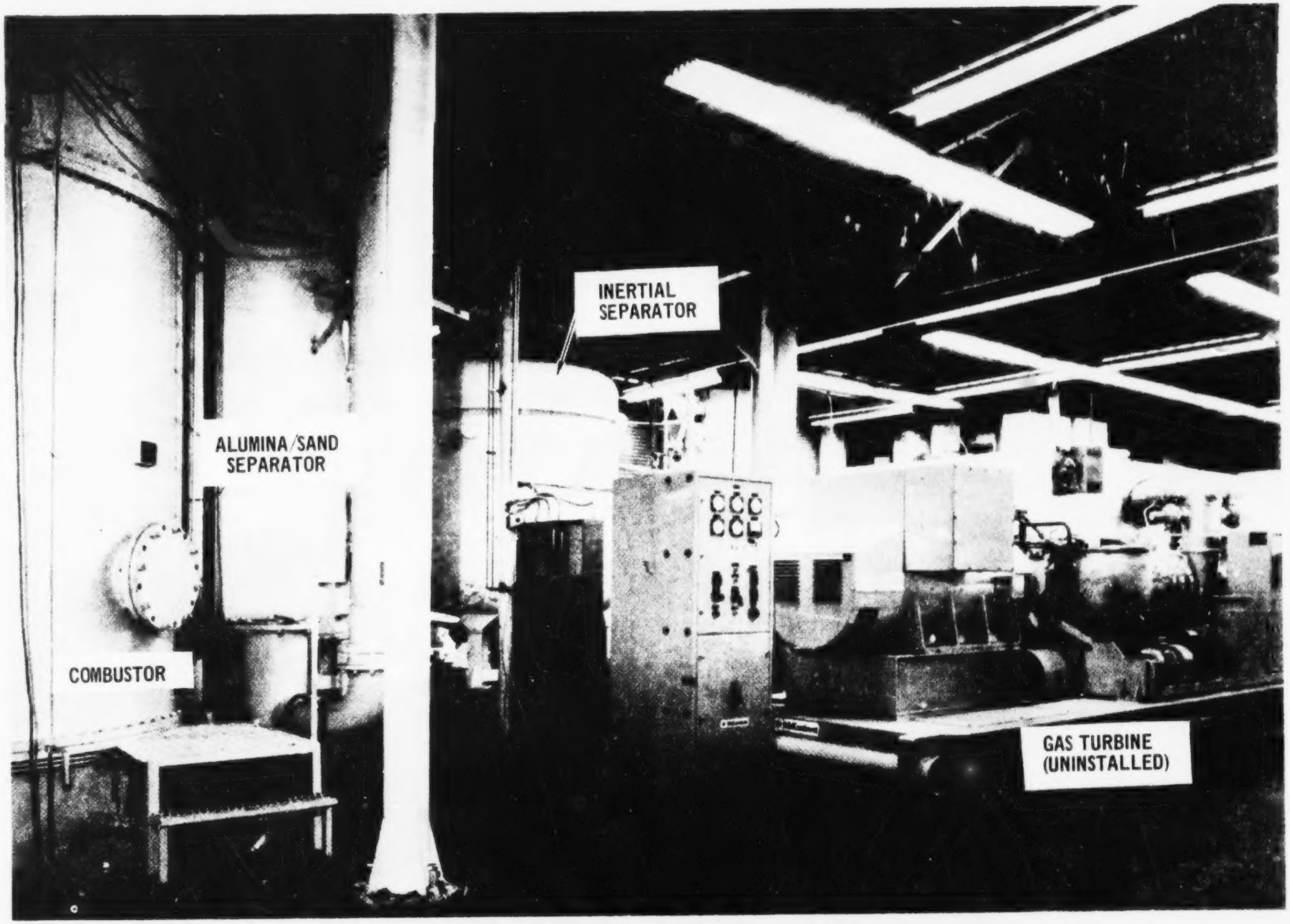

Figure 7. View of solid waste combustor and gas preparation subsystem.

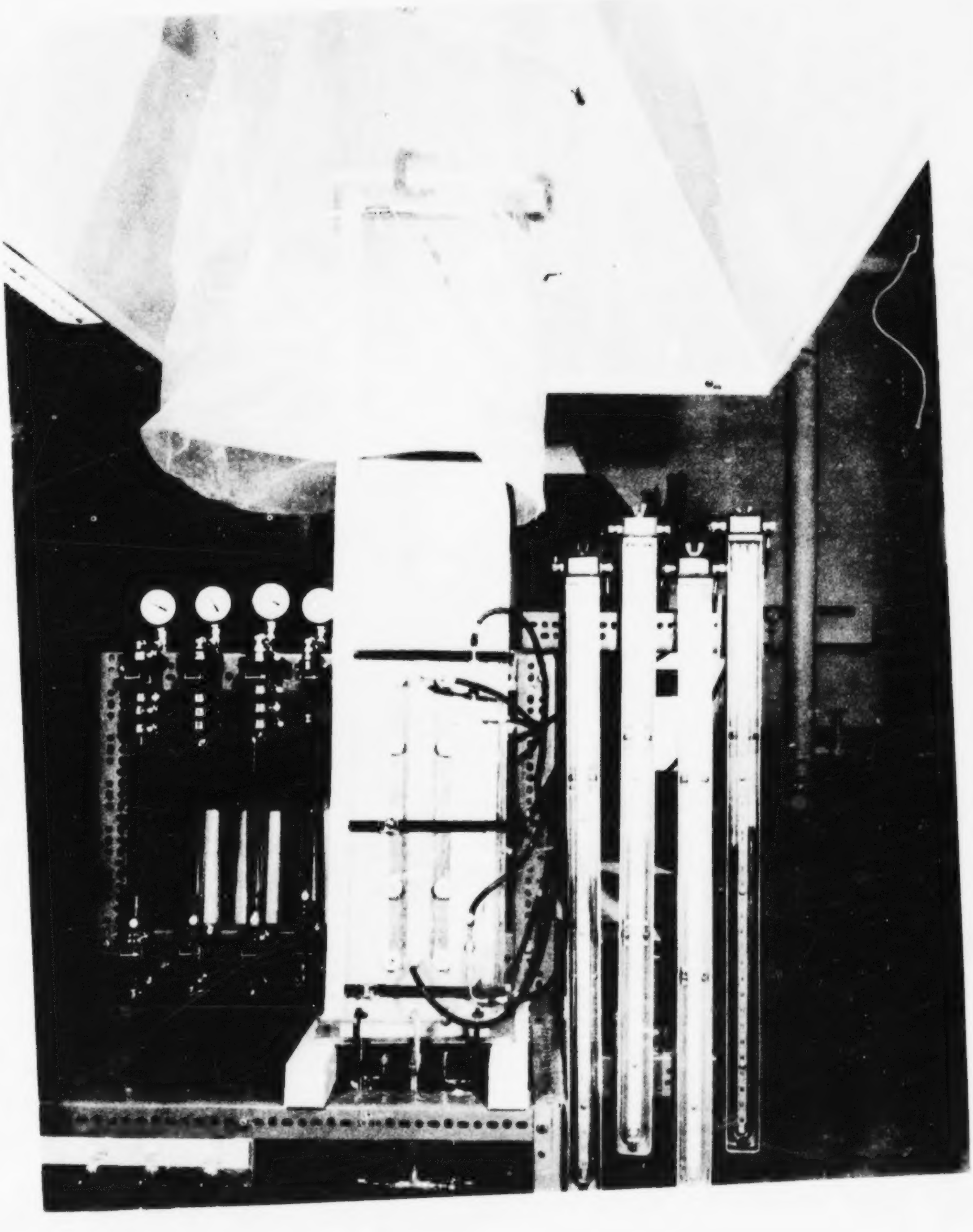

Figure 8. Two-dimensional cold model recirculating bed.

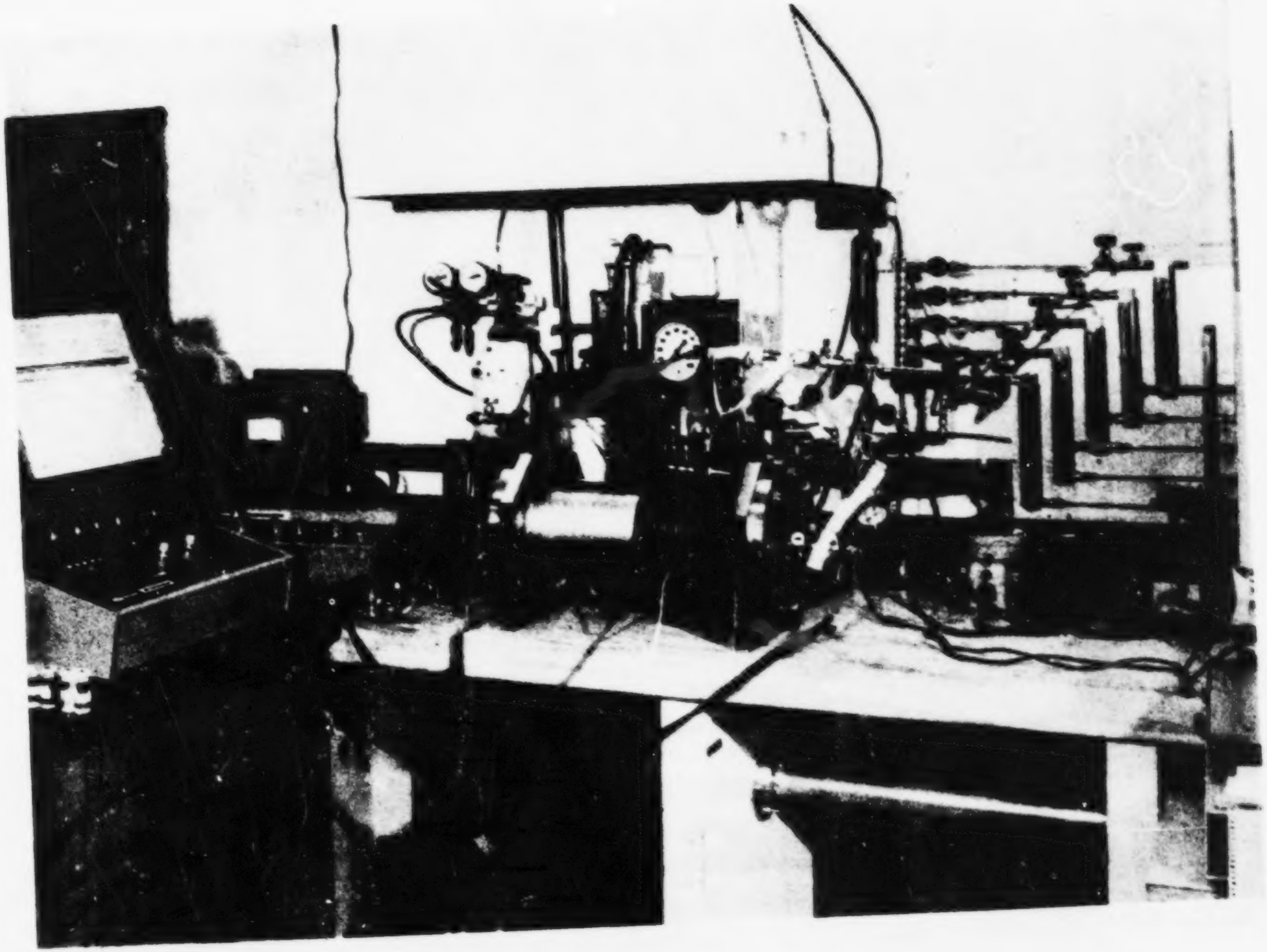

Figure 9. Thermogravimetric analyzer for high temperature and pressure reaction studies on limestone and char.

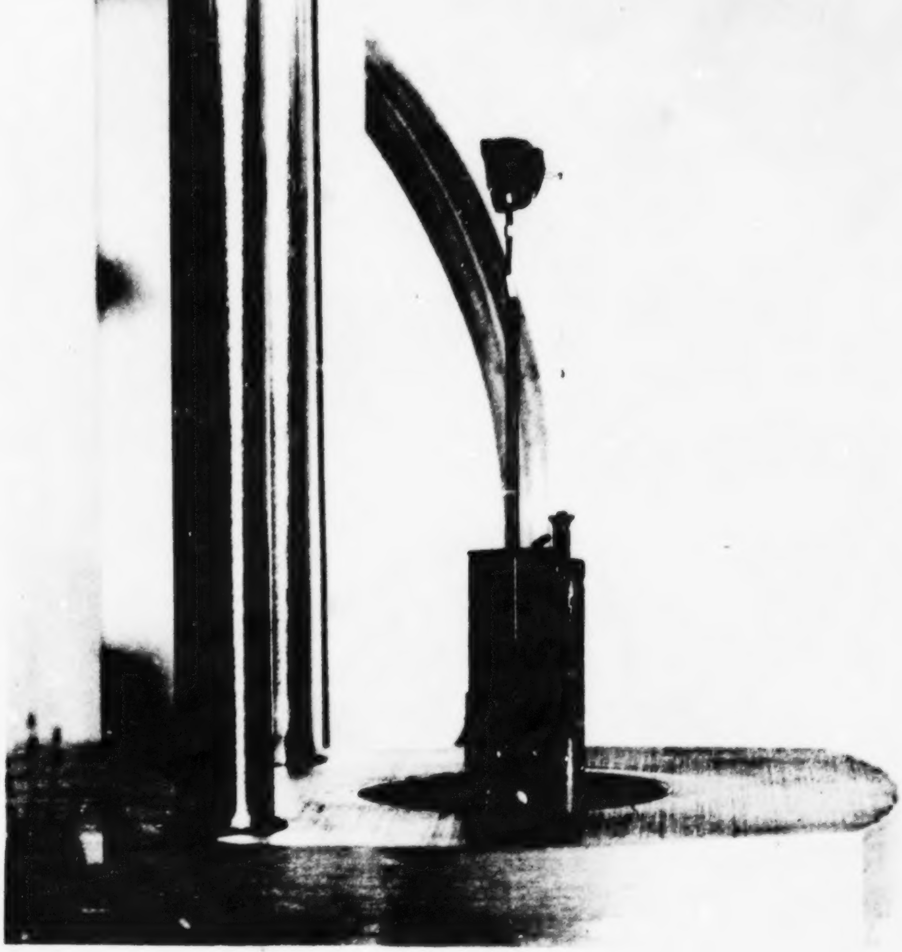

Figure 10. Sample holder and balance arm of thermogravimetric analyzer.

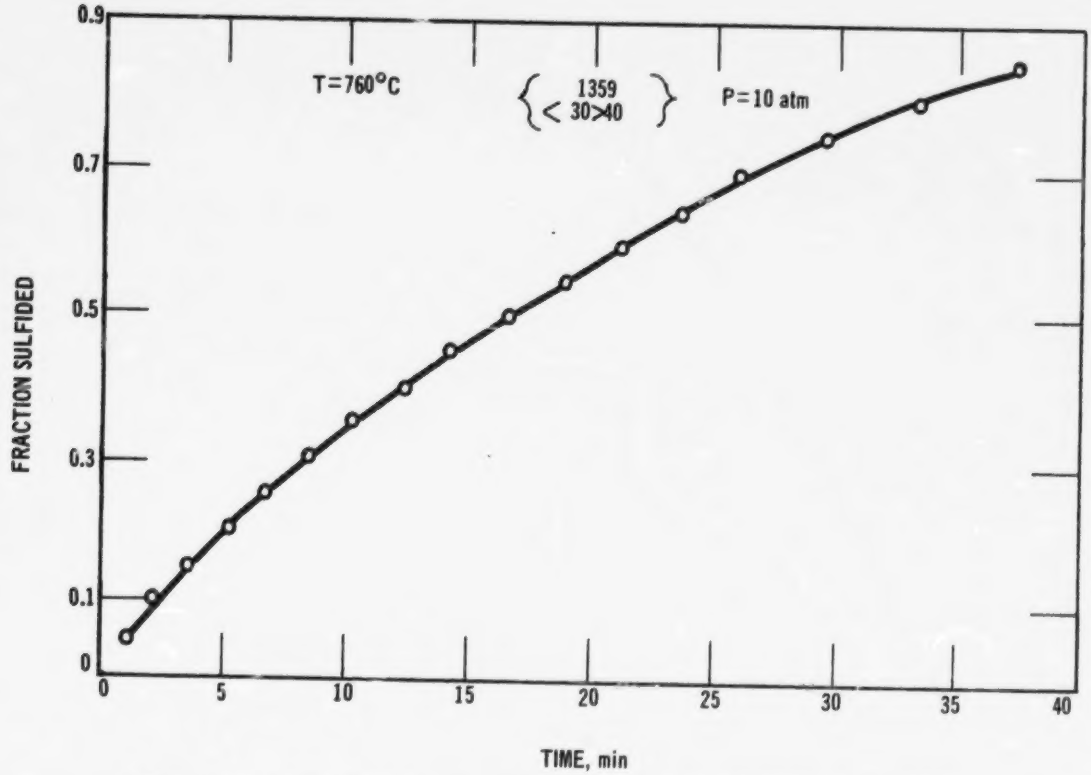

Figure 11. Sulfidation of calcined limestone at $H_2S + CaO \rightarrow CaS + H_2O$.

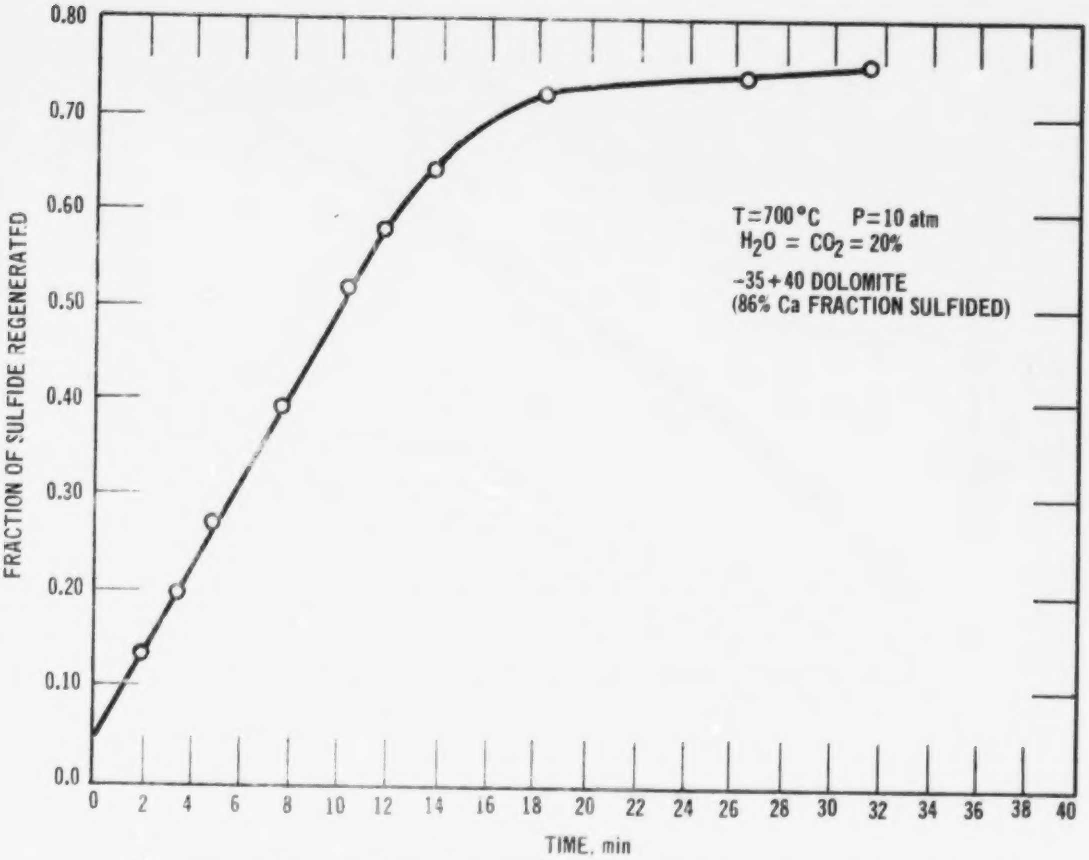

Figure 12. Regeneration of carbonate from sulfided dolomite $CaS + H_2O + CO_2 \rightarrow CaCO_3 + H_2S$.

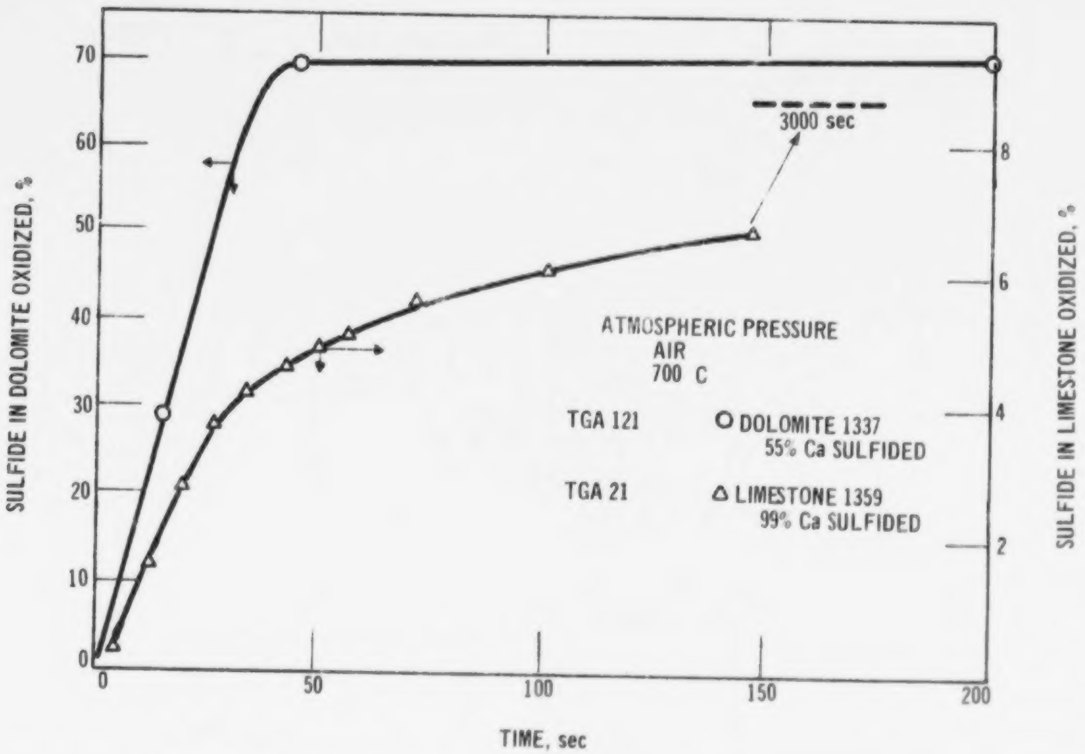

Figure 13. Oxidation of sulfided limestone $CaS + 2O_2 \rightarrow CaSO_4$.

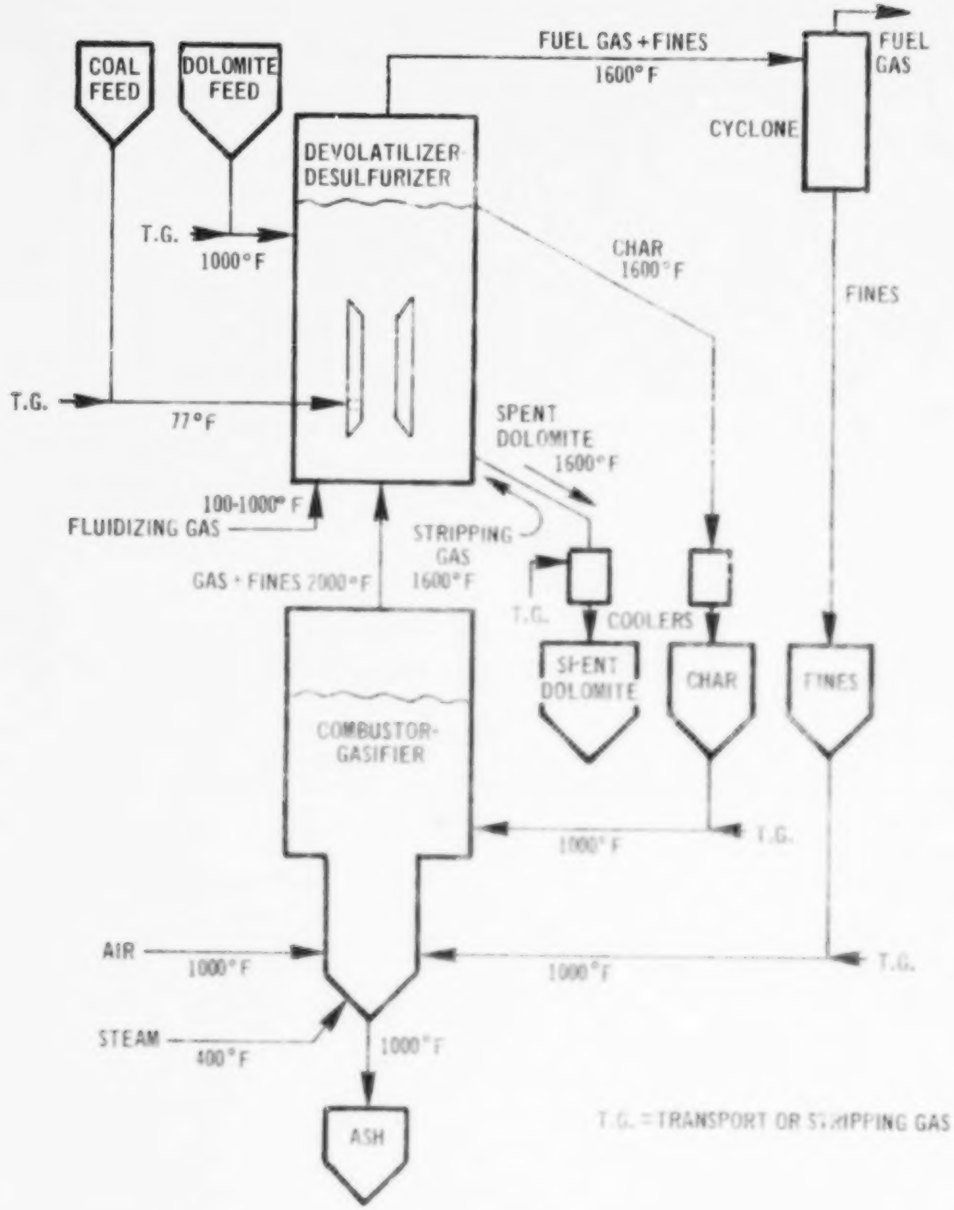

Figure A1. Flow diagram for 1200 lb/hr process development plant.

4. SULFUR RETENTION IN FLUIDISED BEDS OF LIME UNDER REDUCING CONDITIONS

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ABSTRACT

Work done at the Esso Research Centre at Abingdon under EPA. OAP Contract CPA 70-46 has amply confirmed the early promise of the oil-fired fluidised-bed desulphurising gasifier. Since the Second International Conference on Fluidised-Bed Combustion an extensive test programme has been carried out on a batch basis. In addition, the 7 x 10^6 Btu/hr continuously regenerating gasifier, which was previously described, has been constructed and successfully operated. The results show that the gasifier will operate satisfactorily at lower stoichiometric ratios and at higher temperatures than were previously established.

INTRODUCTION

Papers presented at the First and Second International Conferences on Fluidised-Bed Combustion dealt with the mechanisms of sulphur absorption in fluidised beds of lime, under oxidising and reducing conditions. Information was also given for the gasifying case, concerning the effect on desulphurising efficiency of variations in stoichiometric ratio, the mean particle size of the bed material, and bed replacement rate in cyclic operation. Work carried out under OAP contract CPA 70-46 has enabled this information to be supplemented considerably. In this more recent work an American limestone, BCR 1691, was used in conjunction with a western hemisphere fuel oil.

Batch Test Equipment

Two batch gasifiers were specially constructed for the programme but were essentially similar to those used previously. Figure 1 shows a drawing of the new reactor vessel with its various connections. The reactor is made of mild steel lined with a castable refractory. The lower section containing the fluid bed is 7 inches in diameter and 33 inches high. Fuel oil enters the reactor through a single 1/4-in. diameter nozzle which protrudes 1 inch from the reactor wall at a point 5 1/2 inches above the bottom. The upper, disengaging, section of the reactor contains two cyclones which can be drained externally. The distributor is of radial form cast in refractory cement, with 16 horizontal holes distributed around its circumference. The units are brought into operation by underfiring with gas and kerosine. When gasifying, product gas leaves through a cyclone outlet and is flared outside the laboratory. A portion of the gas is burned in a sample flare located above the reactor; the combustion products are analysed for SO_2, O_2, CO, and CO_2. Because of the wide range of sulphur compounds which might be present in the product gas itself, this is the only practical method for measuring desulphurising efficiency. During regeneration similar analyses are made of the undiluted gas.

Batch Test Methods

Two types of tests were made, fresh bed tests and cyclic tests. A new batch of calcined lime was used in each of the fresh bed gasification tests. These tests were used to rapidly screen the effects of the following variables: bed depth, gas velocity, particle size, and air/fuel ratio. The cyclic tests are the nearest simulation to continuous gasifier operation that can be obtained in batch units. The same charge of lime is subjected to repeated cycles of sulphur adsorption and regeneration. After each regeneration a portion of lime is removed and is replaced by an equivalent amount of fresh limestone. The added limestone is calcined to lime during the early part of the next gasification cycle. Without this replacement the activity of the bed gradually declines. With replacement, the activity falls initially, but after a few cycles it reaches an equilibrium level determined by the replacement rate.

Batch Test Results

Because the fuel handling capacity of a gasifier of given dimensions increases as the air/fuel ratio decreases, efforts have been made to operate at the lowest possible air/fuel ratios and to define the limitations that may exist. Under adiabatic conditions there is a relationship between air/fuel ratio and operating temperature. In the batch units, however, the operating temperature was in any case lower than the adiabatic level because of heat losses through the walls and could be lowered still further by means of the bed cooling heat exchanger.

The results plotted in Figure 2 show how changes in operating temperature affect desulphurising efficiency over a range of air/fuel ratios. All of these tests were made using different batches of the same bed material and in each case there was 5 percent by weight of sulphur in the bed material when the plotted result was obtained. The figures in parentheses by each point indicate the carbon content of the bed material. It can be seen that there is a tendency for desulphurising efficiency to fall as the air/fuel ratio is lowered, and that this tendency is increased when the operating temperature is lowered from between 840 and 870°C to 800°C. It can also be seen that there is, as would be expected, a tendency for the carbon content of the stone to rise as the air/fuel ratio is lowered; this tendency, too, is increased when the operating temperature is lowered. It would not seem unreasonable to infer that the presence of a surface layer of carbon reduces the reactivity of the stone. Further evidence supporting this view is shown in Figure 3. In this case desulphurising efficiency is plotted against percent of calcium utilisation at four different air/fuel ratios. The figures in parentheses again relate to the carbon content of the stone at the adjacent point. If the point at 0.38 percent by weight of carbon is compared with the point at 4.68 percent by weight of carbon, it will be seen that although the temperature was about the same in the two cases, 850 as against 845°C, the desulphurising efficiency at the higher carbon content was much lower. The results obtained at an air/fuel ratio of 14.8 percent of stoichiometric and a temperature of 780°C might also have been affected by some degree of recarbonation of the stone; this might account for the relatively poor desulphurising efficiency at very low calcium utilisations when the carbon content was also probably quite low.

At this point it might be appropriate to discuss the factors controlling stone carbon content. It has been found that much more carbon than hydrogen is oxidised in the gasifier. The injected oil cracks on the surface of the stone laying down carbon which is oxidised when the stone reaches the vicinity of the distributor. It follows that the amount of carbon on the stone is a function of the rates of deposition and removal; the rate of deposition reflects the rate at which fuel is injected, and the rate of removal reflects the availability of oxygen and the relative proportions of CO_2 and CO which are produced.

It is evident that CO_2 is the predominant combustion product which is produced in the vicinity of the distributor where the incoming air meets the carbon coated bed material. As the combustion products pass up through the bed, however, there is a tendency for the CO_2 to be reduced to CO at a rate dependent upon the temperature and the availability of carbon. This point is aptly illustrated by the data plotted in Figure 4. These data relating CO/CO_2 ratio with temperature were obtained during the regeneration of a bed which initially contained 7 percent by weight of carbon. Readings of temperature and gas composition were taken at one minute intervals, and no oxygen appeared during the period under consideration. It may be seen that during the first two intervals there was a steep increase in CO/CO_2 ratio as the temperature rose. Subsequently, however, as the carbon content of the bed fell so did the CO/CO_2 ratio despite a further rise in temperature. As a matter of interest, the thermodynamic equilibrium gives a CO/CO_2 ratio of 40 at 900°C.

An example of the working of this mechanism during gasification is given by the data plotted in Figure 5. In this figure, percent weight of carbon on lime is plotted against duration of exposure to gasifying conditions. The bottom curve lines out at a very low carbon content within the first 60 minutes of operation, whereas the top two curves show a progressive increase in carbon content over the first 120 minutes; though both of these curves show a tendency for the carbon content to reach an equilibrium level. The figures in parentheses are the desulphurising effeciencies at 120 minutes gasification time; the 865°C run gave a much better result than the 845°C run, although the stoichiometric ratios as well as the temperatures were very close to each other, being 24.8 and 24.1 percent. It seems reasonable to attribute this difference in performance to the difference in carbon content, these being 0.2 and 6.0 percent by weight, respectively. It may be deduced from these results that it is advantageous to run the gasifier at as high a temperature as possible at low stoichiometric ratios. Unfortunately, due to the heat loss through the walls of the small batch reactors it has not been possible to approach adiabatic conditions; but experience with the continuously operating gasifier has confirmed that good results are obtainable at air/fuel ratios in the region of 18 percent of stoichiometric and temperatures in the region of 870°C. This is an area which will be explored in more detail.

The tests which gave the results which have so far been discussed were made in beds 15-in. deep and at superficial gas velocities in the region of 4 ft/sec. In this work, the practice has been to discuss gas velocity in terms of the superficial air rate, i.e., the velocity which the air supplied to the reactor would reach at the operating temperature when flowing through the empty vessel. The actual superficial gas velocity is higher, due to the presence of cracked oil products; the deviation will increase as the air/fuel ratio falls.

The curve plotted in Figure 6 shows the basic effect of variations in superficial gas velocity. In these tests the bed depth was 15 inches, the operating temperature was 870°C, the air/fuel ratio was 25 percent of stoichiometric, and there was 5 percent by weight of sulphur in the bed material. These data were obtained using a U.S. fuel and fresh beds of U.S. stone, BCR 1691. It can be seen that satisfactory results were obtained at superficial gas velocities up to 6 ft/sec, but at 8 ft/sec there was a marked deterioration in performance. The next step was to check the effect of varying the bed depth. Figure 7 shows the results which were obtained at a gas velocity of 6 ft/sec, with all variables other than bed depth held at the levels used in the previous set of tests. In this case satisfactory results were obtained at bed depths greater than 15 inches but a marked fall in desulphurising efficiency occurred when a 10 in. bed was used. At a gas velocity of 8 ft/sec the desulphurising efficiency was only 40 percent with a 10-in. bed, but reached nearly 100 with a 20-in. bed.

The size of the stone which was used to obtain these results was in the range 300 to 3000 μm. This size range is obtained by dedusting the 1/8-in. diameter tailings from a normal limestone quarry screening operation. As a matter of interest some comparative tests were made with narrower size range fractions sieved from this stone. The results of these tests are shown in Figure 8. It can be seen that the narrower sized fractions gave markedly poorer results than the material from which they were obtained. The results listed in Table 1, however, show that when the smaller of the two narrow cut fractions was substituted for the make-up feed of a full size range bed under cyclic conditions, contrary to expectations, the desulphurising performance was improved. There is no clear-cut explanation for this effect, but it is possible that the single cycle results reflected quality of fluidisation as well as particle size.

Table 1. EFFECT OF LIMESTONE PARTICLE SIZE ON DESULPHURIZING EFFICIENCY IN CYCLIC TEST

	Test number	
	T-3 (Cycles 1-14)	T-3 (Cycles 24-31)
Size range of makeup, μm	300 to 3175	600 to 1400
Replacement rate CaO/wts	2.38	2.37
Lined out SRE, %	61	68
Sulfur Input	3.0	3.1

In the case of fresh bed tests, the extent to which the stone is reacted is not important; but when cyclic tests are made it is necessary to choose realistic levels. It is possible to deduce on thermodynamic grounds that under continuously operating adiabatic conditions the reaction in the sulphur content of the stone per pass through the regenerator will be in the region of 1.0 to 1.5 percent by weight. It follows that it is not possible to operate a cyclic test, in the absence of progressive coking, which adds more fuel, unless a sulphur content higher than about 2 percent by weight is obtained at the end of each absorption cycle. This somewhat higher sulphur content is required because of the additional heat demand which occurs in a batch regeneration. In continuous operation, more or less adiabatic conditions obtain; in a batch operation the refractory lining must be raised in temperature during regeneration as well as the bed, and the heat losses through the wall of the reactor are appreciable. Because a fair amount of oxidation occurs at relatively low temperatures there is a tendency for more calcium sulphate to be formed during batch regenerations than would be expected under continuous conditions. This sets the level of the sulphur content of the bed at the beginning of the subsequent test. This level is generally found to be about 2 percent by weight.

The fuel which was used for the cyclic tests had a sulphur content of 2.3 percent by weight; for convenience, a standardised run duration of 45 minutes was generally used. When account is taken of the range of gas velocities and stoichiometric ratios which were covered this gave sulphur inputs ranging from 1.5-3.7 percent by weight on lime per run.

The first series of cyclic tests were aimed at obtaining a direct comparison of the reactivities of BCR 1691 and the U.K. stone which had previously been tested. The U.K. stone is about 98 percent $CaCO_3$, whereas BCR 1691 is of inferior quality, containing only 88 percent $CaCO_3$. Fresh bed tests had given results indicating that the two stones were equally effective when reacted to the same degree. Cyclic tests, however, revealed that BCR 1691 is so inferior in performance under these conditions that it requires about three times the stone replacement rate for an equal desulphurising performance in a bed 15.5-in. deep. Fortunately, it was also found that a modest increase in the depth of the bed gave an improvement in performance which was quite disproportionate, allowing the stone replacement rate to be substantially reduced. An indication of the nature of the relationship between bed depth, stone replacement rate, and desulphurising efficiency is given by the data shown in Table 2. All of these tests were made at 870°C with 25 percent of stoichiometric air and a superficial gas velocity of 6 ft/sec.

Table2. EFFECT OF BED DEPTH AND STONE REPLACEMENT IN RATE DESULPHURIZING EFFICIENCY IN CYCLIC TESTS

Bed depth, inches	Moles CaO per moles S	Lined out S.R.E., %
15.5	1.39	61
20.0	1.43	90
20.0	2.9	98
22.0	1.4	97

It will be seen that increasing the bed depth from 15.5 to 20 inches improved the desulphurisation efficiency from 61 to 90 percent at roughly the same stone replacement rate of approximately 1.4 mole CaO/mole sulphur. In order to improve the desulphurisation efficiency to 98 percent with a bed depth of 20 inches, it was necessary to increase the stone replacement rate to 2.9 mole CaO/mole sulphur. When, however, the bed depth was increased to 22 inches then a 1.4 mole CaO/mole sulphur stone replacement rate gave a desulphurisation efficiency of 97 percent. It remains to be seen whether BCR 1691 will give a similar performance in the continuous gasifier at a superficial gas velocity of 6 ft/sec.

Results Obtained Operating the Continuous Gasifier

The continuously operating gasifier has been described previously; a detailed discussion of its construction and commissioning is not within the scope of this paper. A general view of the installation, however, is shown in Figure 9. During the commissioning period three runs were made giving a total operating time under gasifying conditions of about 460 hours. U.K. stone was used in these tests, but with U.S. fuel containing 2.5 percent by weight of sulphur and 350 ppm of vanadium. The prime purpose of these runs was to demonstrate fuel gasification with sulphur removal on a continuous basis; the study also took a quick look at the effects of some of the controllable variables. The study showed that the gasified fuel ignites readily and burns with a bright luminous stable flame. Smoke free operation was obtained with about 1.5 percent oxygen in the flue gas over long periods. This is a better performance than that given by the conventional oil burner which the gasifier replaced. When tested prior to conversion it was found that the package boiler required about 3 percent oxygen in the flue gas for smokeless operation.

The operating conditions covered during the test runs are indicated in Table 3. In general the superficial gas velocity was about 4 ft/sec; when the fuel rate was varied the operating temperature was controlled by recycling flue gas. During a considerable proportion of the operating time desulphurisation was virtually complete. Although this result was highly gratifying, it did not yield much information concerning the effects of the independent variables. In one period of 19 hours duration in which virtually no SO_2 was detected in the flue gas, the running conditions were as shown in Table 4. The gasifier temperature was about 900°C, the pressure drop through the bed averaged 14.5 inches water gauge the air/fuel ratio was 23 percent of stoichiometric, and the stone replacement rate was about 1.4 mole CaO/mole sulphur entering the bed. The superficial gas velocity averaged 3.7 ft/sec. In another period of 25 hours duration at the end of the test, the operating temperature was about 880°C, the pressure drop through the bed was about 19.5 inches water gauge the air/fuel ratio was 22 percent of stoichiometric, and the desulphurisation efficiency averaged 95 percent. During the first 16 hours of this period the stone replacement rate was about 1 mole CaO/mole sulphur, during the remaining 9 hours the Ca/S ratio was 0.6 of stoichiometric giving an average figure for the 25 hours of 0.85 mole CaO/mole sulphur. In the last hour the air/fuel ratio fell to 18 percent of stoichiometric, but the desulphurising efficiency never fell below 91 percent.

Table 3. CONTINUOUS GASIFIER OPERATING CONDITIONS

Programme item	Pilot plant operation
Number of test	3
Test duration	91-202 hr
Limestone used	U.K., Denbighshire
Oil used	Venezuelan 2.5% S
Gasifier temperature	820-920 °C
Regenerator temperature	1050-1100 °C
Bed depth	13-23 in.
Superficial gas velocity	2.8-4.3 ft/sec
Lime particle size range	300/3200-800-3200 μm
Lime replacement rate	0.54-4.8 mole CaO/moleS
Air fuel ratio	15-31% of stoichiometric
Oil feed rate	61-82 lb/hr-ft^2

Table 4. GASIFIER PERFORMANCE

Duration of experiment, hr	19	25
Gasifier temperature, °C	900	880
Air/fuel ratio, % stoichiometric	23	22
Bed pressure drop, in. H_2O	14.5	19.5
Gas velocity, ft/sec	3.7	3.9
Stone replacement rate, moles CaO/moles S	1.4	0.85
Sulphur removal efficiency, %	100	95

The operational problems which were encountered during these runs, which totalled 460 hours under gasifying conditions, were of a minor nature and remedial action has since been taken. In view of the results which were obtained there is no doubt at all that the process is a feasible proposition.

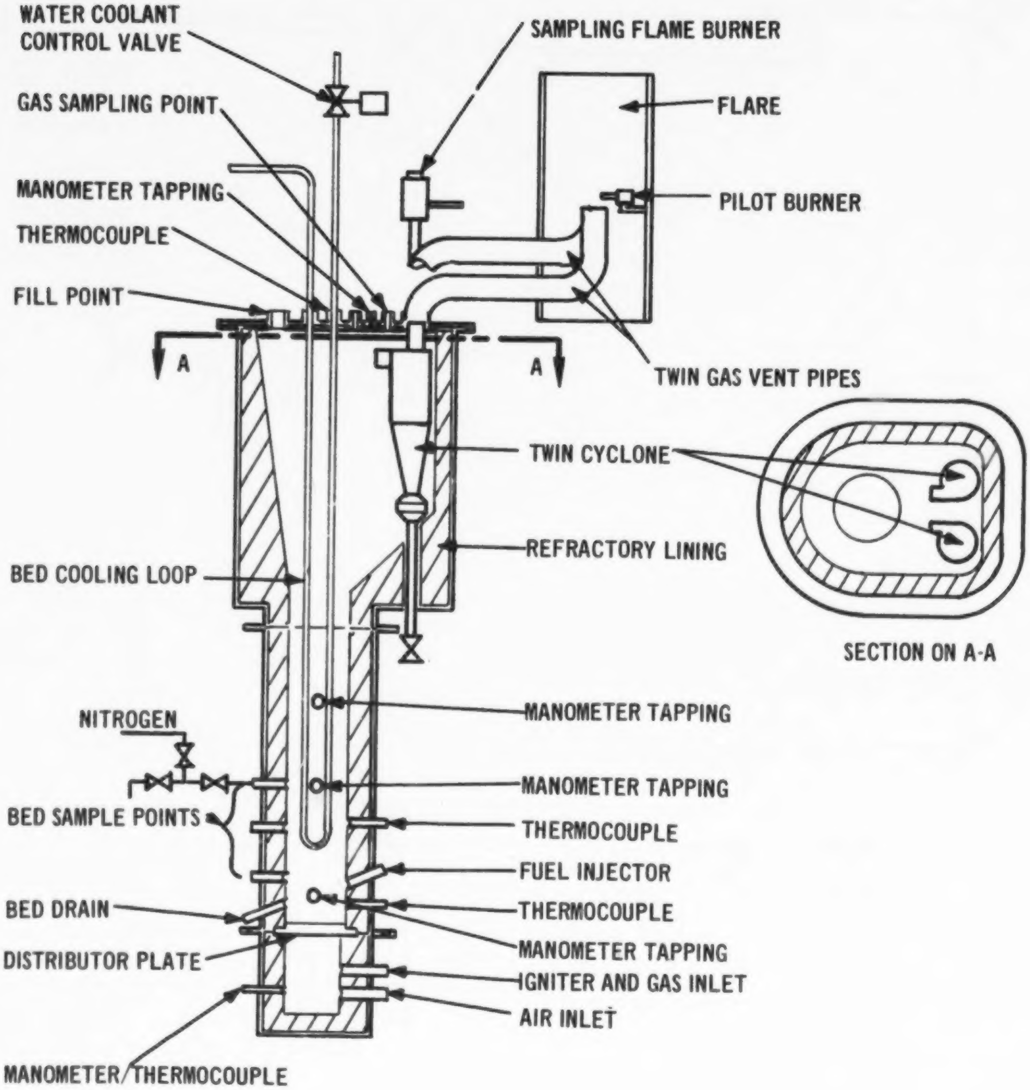

Figure 1. CAFB batch unit reactor.

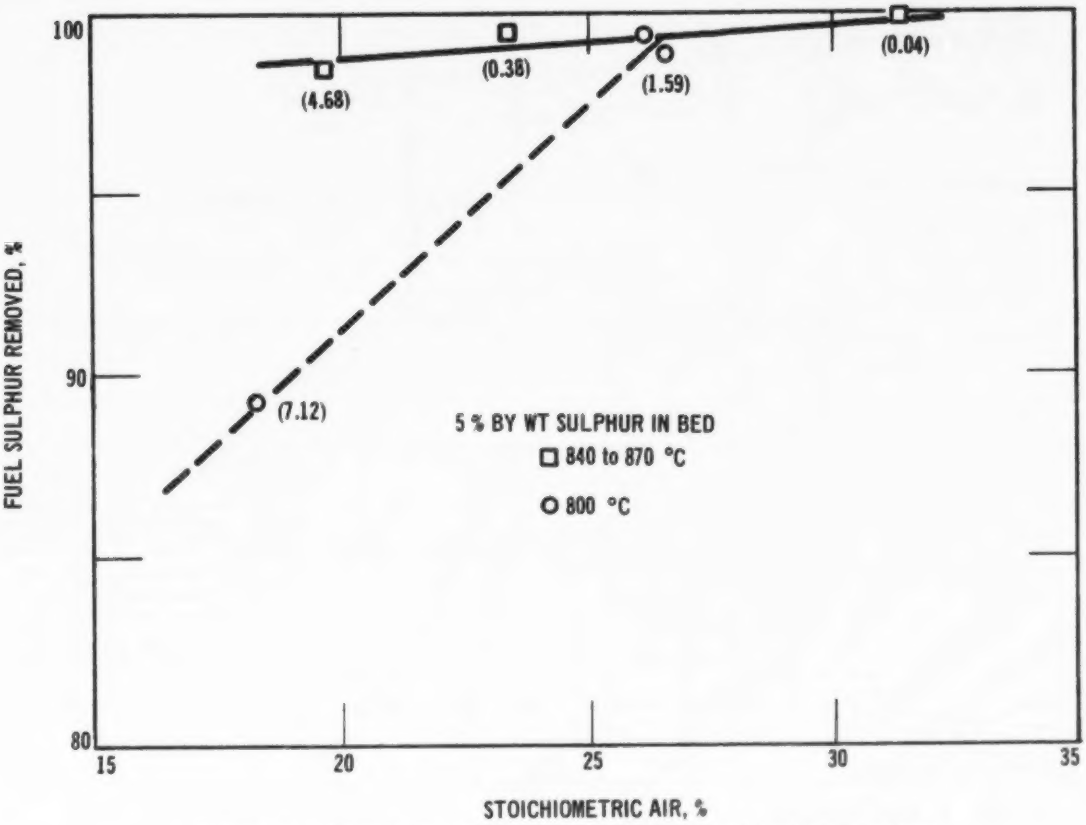

Figure 2. Interaction between air/fuel ratio and bed temperature.

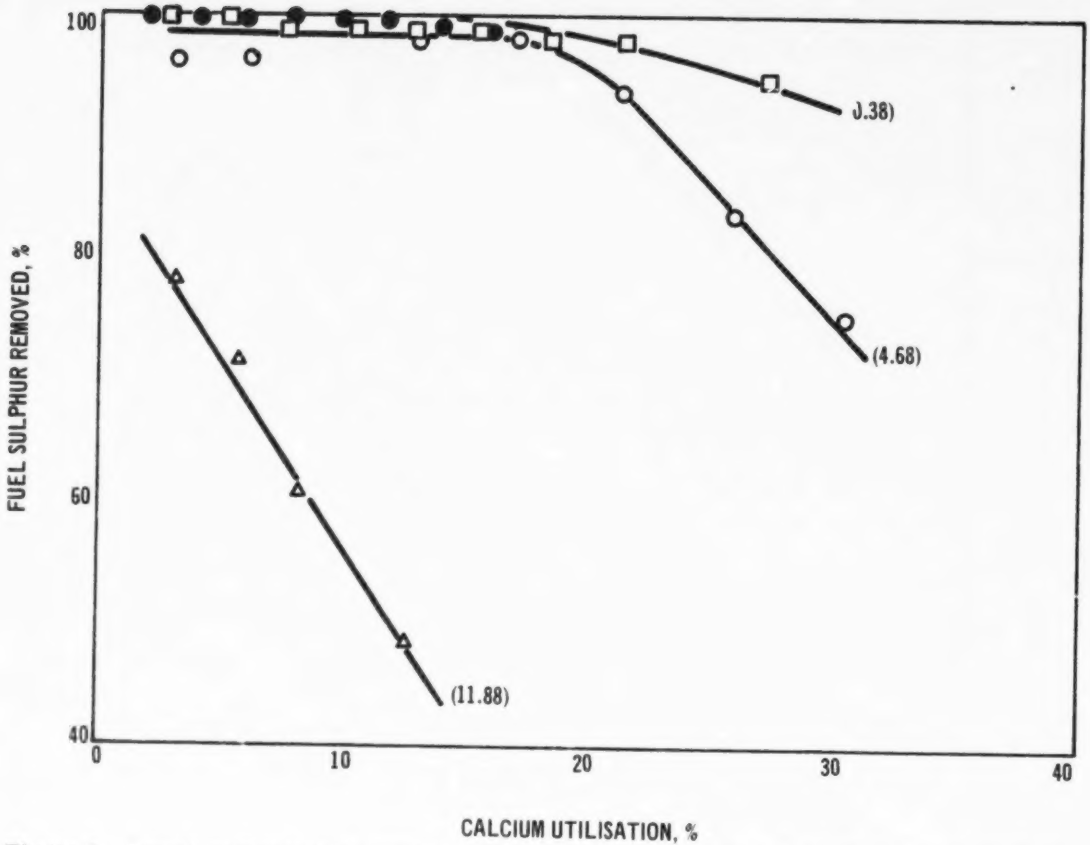

Figure 3. Result at 6 ft/sec in number 4 units (sulphur removal curves at different air/fuel ratios).

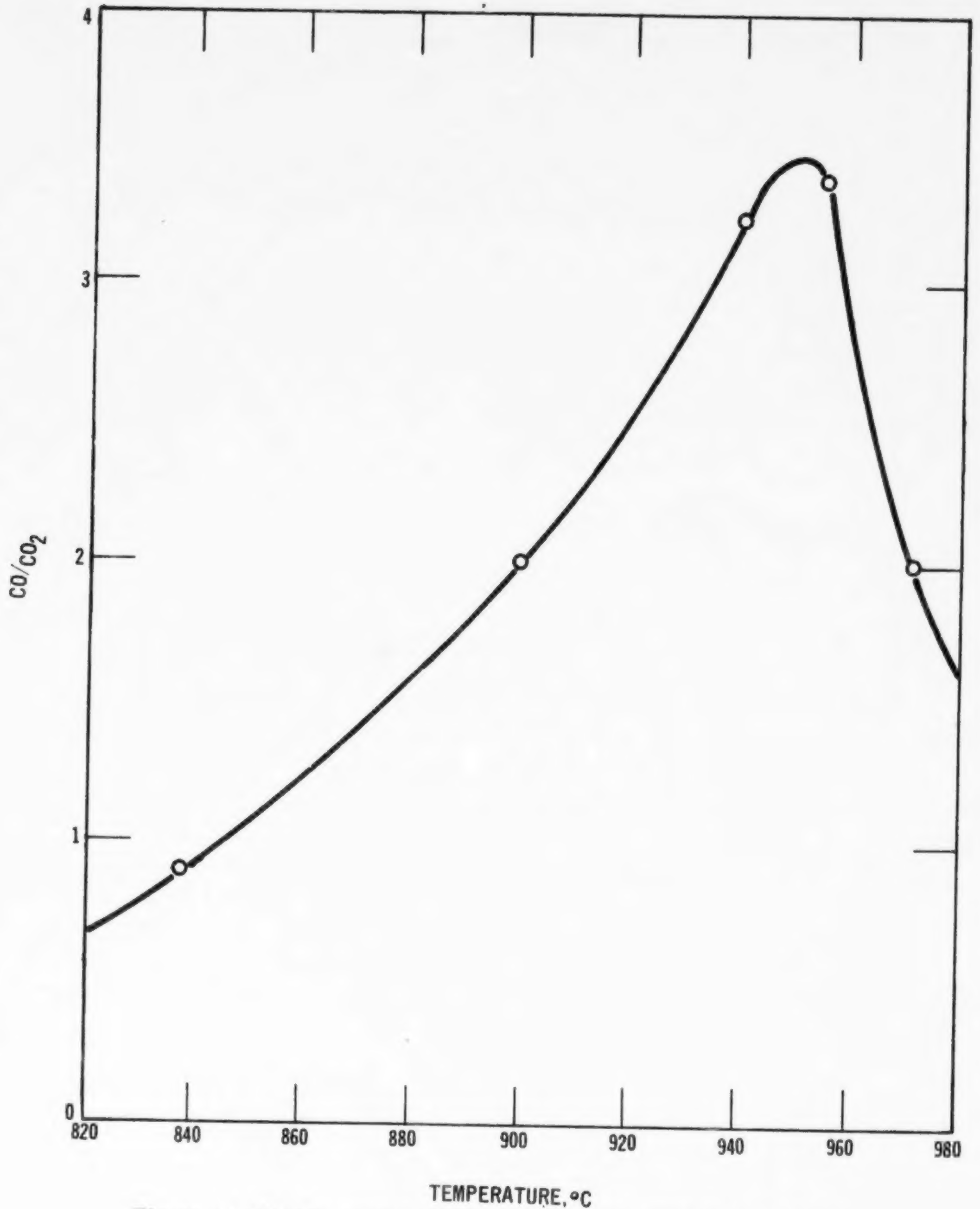

Figure 4. CO/CO$_2$ profile during regeneration, fresh bed test no. 7.

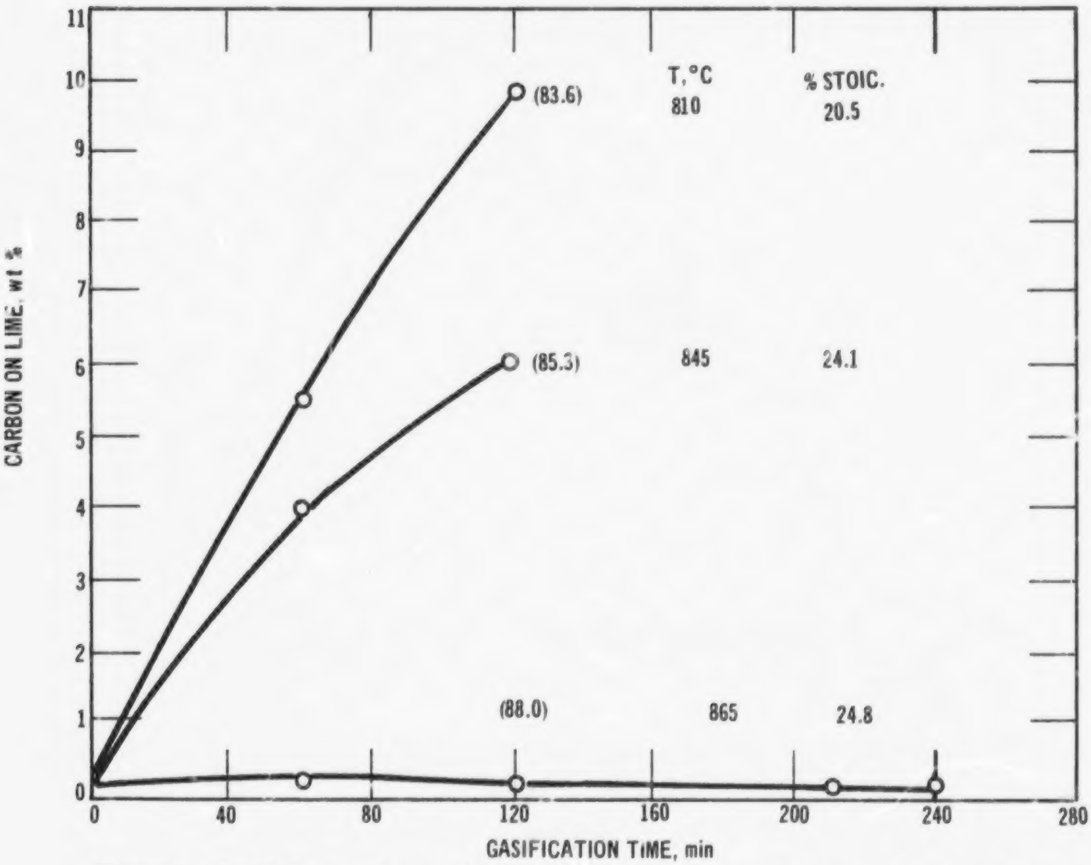

Figure 5. Change in carbon content of lime during batch gasification cycle 1-A unit.

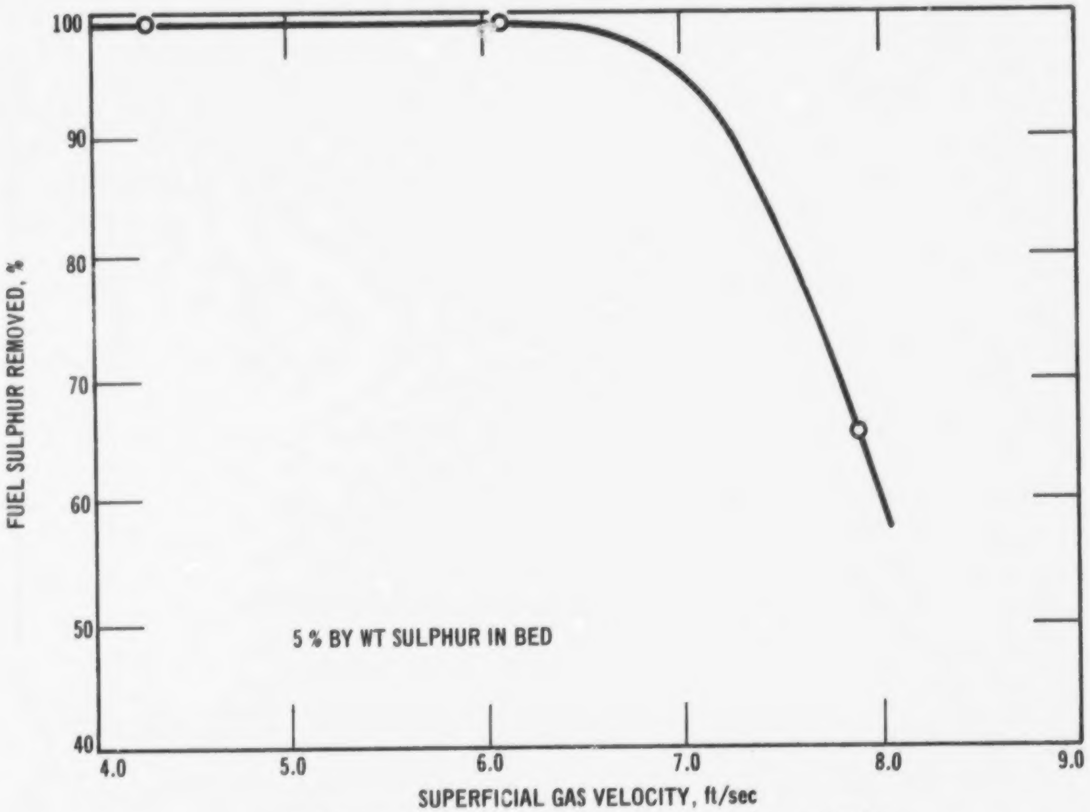

Figure 6. Basic effect of superficial gas velocity.

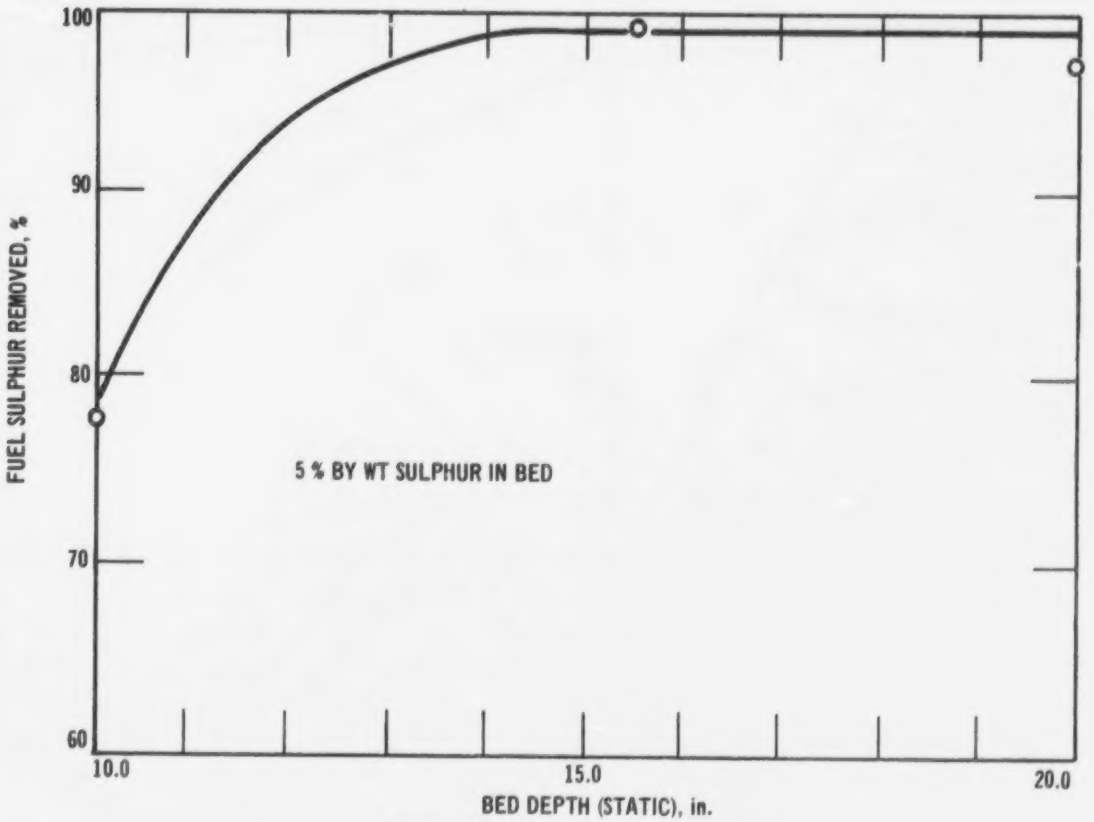

Figure 7. Basic effect of bed depth.

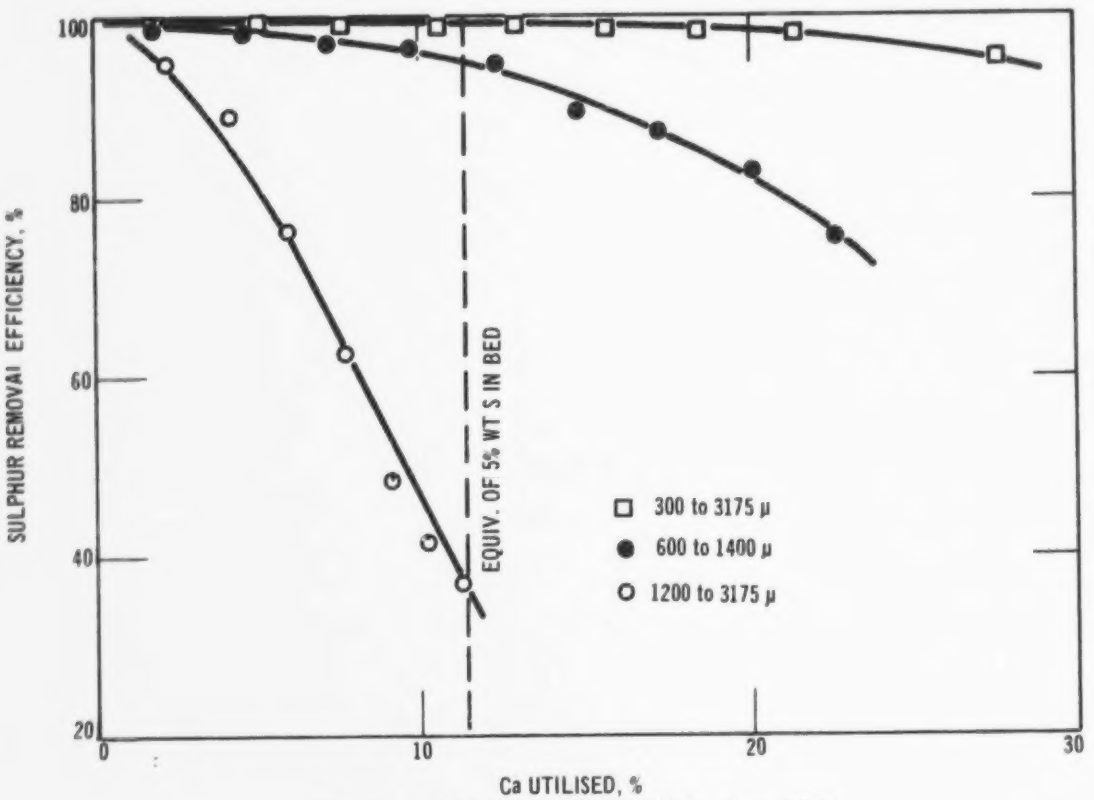

Figure 8. Basic effect on particle size range.

5. THE CO_2 ACCEPTOR GASIFICATION PROCESS—A STATUS REPORT—APPLICATION TO BITUMINOUS COAL

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ABSTRACT

This paper discusses experience gained and problems encountered during startup operations of the Rapid City pilot plant. A project schedule is given for completion of that phase of the work aimed at production of pipeline gas from low-rank western coals.

Process revisions that must be made in application of the CO_2 acceptor system to high-sulfur bituminous coals are discussed. The major revisions are installation of pretreatment facilities to handle caking coals and an increase in gasification temperature to accommodate the poorer reaction kinetics.

Experimental work on the pretreatment of bituminous coals to render them suitable for pressurized gasification by preoxidation. Highly fluid coals such as Pittsburgh seam do not. Promising results are reported via a "Seeded Coal" type process.

A revised flow sheet and heat and material balance is given for application of the CO_2 acceptor process to the processing of bituminous coals. Recycle of CO_2 is a key feature in this operation.

INTRODUCTION

The CO_2 acceptor process has been under development for a number of years. The major goal of this work has been the production of pipeline gas from low-rank western coals. The process has been extensively described in numerous previous publications and no description is deemed necessary here. A relatively complete description of the technical basis of the process and its economic potential is available in reports to the Office of Coal Research.

A pilot plant to test the process has been constructed at Rapid City, South Dakota. The project is financed jointly by the Office of Coal Research and the American Gas Association.

The purpose of this paper is to give a brief status report on the Rapid City operations and of the contemplated development schedule. It also discusses problems and opportunities involved in the application of the process to treatment of bituminous coal. The use of bituminous coals in the process is not only of interest for production of pipeline gas, but more broadly for the production of low-sulfur boiler fuel.

The production of low-sulfur boiler fuel from bituminous coals by an adaptation of the process to produce a low-Btu gas without[2] and with low-sulfur char[3] as co-product has been studied in the course of a research contract between Consolidation Coal and the EPA. This work is discussed in more detail in a succeeding paper at this conference. It also formed the subject of a paper presented at the Second International Conference on Fluidized-Bed Combustion. This paper gives a brief resume of some work now being conducted for the EPA in the pretreatment of caking bituminous coals to establish operability in pressurized fluid-bed gasifiers.

STATUS OF PILOT PLANT DEVELOPMENT

The construction of the pilot plant was completed with formal acceptance of the plant on December 28, 1971. Mechanical testing of the various plant components occupied the next period through the end of March 1972. During this test period, a number of unit operations were successfully carried out. These included operation of the gas purification, char grinding, and lockhopper systems. The fired heaters were put on stream and the process vessels were successfully heated to 1400-1600°F by hot gas circulation.

Pilot plant operations since April 1972 were aimed at initiation of an actual gasification run using lignite char as a feedstock. The initial run was chosen to demonstrate a simplified two vessel version of the process to be conducted at 150 psig. The system to be demonstrated is illustrated by two vessels shown in Figure 1 — the gasifier and regenerator. The details shown on the remainder of the flow sheet should be disregarded at this time since they refer specifically to future operations with bituminous coal which will be discussed later. It should also be noted that in the pilot plant runs the temperature in the gasifier was programmed for lower temperatures than indicated in Figure 1, i.e., at 1520°F.

One of the unique features, from the engineering point of view, in the CO_2 acceptor process is the dual fluo-solids handling system wherein acceptor, which originally is either dolomite or limestone, is fed to fluidized beds of char. The acceptor particles are bigger and heavier than the char particles and shower down through the char bed. They collect as a separate and segregated fluid bed of acceptor in the boot at the bottom of the gasifier from which the acceptor is recirculated to the regenerator.

The ability to maintain a segregated fluidized acceptor bed reasonably free of char is one of the key elements in achieving a successful demonstration of the process.

This feature has been well demonstrated in the prepilot scale work at Consolidation Coal Company's Research Laboratories, but one of the purposes of the Rapid City pilot plant is to demonstrate that this operation can be successfully scaled up.

Acceptor circulation tests were carried out in April, May, and June 1972 between the gasifier and regenerator preparatory to initiation of the gasification tests. Operations in June were hampered by the June 9 Rapid City flood and its aftermath. Difficulty was encountered in these tests due to chronic plugging of the pressure probes used to control the operation. The plugging was due in part to an inadequate purge gas system.

The situation was rectified by increasing the diameter of the pressure probes from 1/4 in. to 1/2 in., installing a rod out system to break plugs, installing duplicate probes at critical measuring points, and improving the purge gas supply system.

Successful hot continuous circulation of dolomite-based acceptor was demonstrated for a period of 14 hours during the end of this period. The $MgCO_3$ portion of the stone was calcined to MgO and the stone circulated in the "half calcined" condition. The run was terminated involuntarily due to a pressure upset and corresponding loss of pressure balance between the two vessels.

One of the problems encountered during this period was a high rate of attrition of the acceptor in its soft, half-calcined condition. This led to excessive generation of fines which caused plugging difficulties at the entrance to the quench towers.

The softness of the stone is of a transitory nature since it is known from our prepilot experience that it hardens rapidly when it is cycled through the actual process operations which were not attempted during these circulation tests.

During July a successful hot acceptor circulation test was performed, and preparations were made again to start an actual gasification run. Beds of half-calcined acceptor and char were established in the gasifier and regenerator, respectively. Circulation of acceptor through the char bed was initiated, but difficulty was experienced in obtaining a distinct char-acceptor interface in the gasifier boot.

The acceptor-char mixture was transferred via the lift leg to the regenerator. The acceptor was, however, rich in char, and since the lift gas was nearly pure air combustion of the char in the lift line was initiated. This caused development of a hot spot in the line which resulted in its rupture and termination of the operation.

The above incident is not typical of normal operation since the lift gas usually is recycle regenerator offgas substantially free of oxygen. In this case air was present because circulation was started before char combustion in the regenerator was initiated.

A fourth startup was initiated after the ruptured lift line was repaired in August. Difficulty occurred at all times, however, with blockages in the acceptor and char transfer lines. It was obvious from both the temperature and pressure profiles that fluidized beds of acceptor in the gasifier boot and char in the gasifier was not being maintained. In spite of this, sufficient acceptor was transferred through the lift line to establish the regenerator bed, and sufficient char was fed to the regenerator to establish combustion therein. The regenerator was increased to 1700°F. The unit was shut down on August 20 for inspection when the char feed line to the gasifier plugged. This was done to determine the cause for the failure to establish the desired fluid beds in the gasifier.

Inspection of the gasifier revealed that the fluidization difficulty was due to a failure of the refractory in the gasifier boot, particularly at the seam between the head and the gasifier shell, which allowed gas to bypass the solids bed through massive holes and cracks in the refractory.

The gasifier was taken down and a new refractory configuration installed. Specifically, the outer layer of soft insulating refractory was removed from the gasifier boot to be replaced with harder castable refractory. The unit, at the time of this writing, was scheduled to be put back in operation again early in October 1972.

The initial operating difficulties have not been related to fundamental deficiencies in the process itself. It is expected that successful demonstration of the CO_2 acceptor process will be achieved in the Rapid City pilot plant in the near future.

The original schedule has been set back about 6 months, first by some delays in completion of construction and second by the startup difficulties outlined above. A new operating schedule has been drawn up and submitted to the Office of Coal Research for their approval. The new schedule calls for completion of the original pilot plant program in July of 1974. The contemplated program, however, does not encompass testing of Eastern bituminous coals.

The processing of bituminous coals is of interest not only for production of pipeline gas but also for production of low-sulfur boiler fuel. A series of new problems are introduced when the process is adapted to use of caking bituminous coals. Extension of the pilot plant operating period as well as some modification of the equipment would be required to study bituminous coal processing.

PROCESSING OF BITUMINOUS COALS VIA CO_2 ACCEPTOR PROCESS

Pretreatment via Preoxidation

The use of fluidized-bed technology for the gasification of caking coals requires that the feed coal be pretreated to render it non-caking in order to sustain an operable bed. The problem becomes more severe as the operating pressure is increased and may also be a function of hydrogen partial pressure. The specific role of hydrogen partial pressure as distinct from total system pressure in intensifying the pretreatment problem has not been fully defined.

The effect of increasing total pressure is illustrated by the two experimental observations outlined below. A highly caking Pittsburgh seam coal was successfully processed in 1949 in an atmospheric pressure, 1 ton/hr fluid-bed gasification unit without any pretreatment.[5] The above admittedly was accomplished at a relatively low coal throughput rate of 25 lb/hr-ft^2, but the effect of higher rates was not explored.

The other observation was, that in processing non-caking sub-bituminous coals at 20 atmospheres pressure in the hydrodevolatilizer of the CO_2 acceptor process, agglomeration of the bed solids occurred unless the coal feed was pretreated by mild preoxidation.[1C]

The work on the development of the synthane process at the USBM[6] again illustrates the fact that successful operation of a pressurized fluidized-bed gasification process with bituminous coal requires that the feed be pre-treated by preoxidation.

A study of the degree of pretreatment required for pressurized fluid-bed gasification of two types of bituminous coals has been carried out for the EPA. One coal was from the Ireland Mine in northern West Virginia and is typical of the highly fluid, high sulfur Pittsburgh No. 8 seam. The other coal was from the Hillsboro Mine in central Illinois and is typical of the more weakly caking, high sulfur Illinois No. 6 coals. The experimental investigation was carried out in the same 4-in. ID reactor previously used in the development of the CO_2 acceptor process.[1C] The experimental method is described in detail in the Annual Report[2] to the EPA. Only a brief summary of results will be presented here.

The gasification conditions chosen for testing operability of the pretreated coals are those outlined in Table 1 and correspond to conditions selected for adaptation of the CO_2 acceptor process to produce low-Btu gas. Under normal conditions in the CO_2 acceptor process, i.e., for production of pipeline gas, a higher partial pressure of hydrogen prevails and even more severe pretreatment may be required.

The severity of preoxidation conditions required to establish operability for the two coals in the gasifier operated at the conditions cited in Table 1 are given for the case of a 28 x 100 mesh feedstock in Table 2.

Table 1. TYPICAL CONDITIONS USED FOR TESTING OPERABILITY OF PRETREATED COALS IN GASIFIER

Temperature, °F	1700	1700
Pressure, psig	206	206
Feed rate, lb/hr	4.83	5.50
Feedstock	28 x 100 mesh, Pretreated, Hillsboro Coal	20 x 100 mesh, Pretreated, Ireland Coal
Fluidizing velocity, ft/sec	0.34	0.33
Input, scfh		
Steam	77	78
CO_2	38	35
N_2	103	111
Air	105	105
Percent carbon burnoff	55	
Mean particle density, lb/ft^3	43.2	

Table 2. MINIMUM CONDITIONS OF SEVERITY OF PREOXIDATION TO PROVIDE OPERABLE GASIFIER FEEDSTOCK

Coal	Ireland Mine	Hillsboro
Temperature of preoxidation, °F	750	810
Size consist of coal feed	28 x 100 mesh	28 x 100 mesh
Stages of preoxidation	2	1
Oxygen consumed, wt % (referred to raw coal)		
Stage 1	18.6	8.7
Stage 2	9.3	—
Total	27.9	8.7

Two criteria are used to evaluate the impact on economics of the results of the preoxidation tests. The first is the percent preoxidation required as compared with the "adiabatic" quantity, i.e., with the amount of oxidation by heat balance to sustain the reaction at the desired temperature.

This relationship is illustrated in Figure 2. It is readily seen that the amount of preoxidation required for Ireland Mine coal is four times the adiabatic quantity at 750°F. This is a strong economic debit since, in order to carry out this process in practice, large amounts of heat must be removed from the preoxidizer. Somewhat larger amounts of preoxidation are permitted by adiabatic operation if the temperature in the preoxidizer is allowed to rise. However, in the case of Ireland Mine coal, substantially higher preoxidation temperatures are precluded since the preoxidizer itself becomes inoperable.

The demonstrated preoxidation severity required for the Illinois No. 6 coal, however, is only slightly above the adiabatic level. As a matter of fact, lower extents of preoxidation may in fact be permissible in this case since an investigation of the lower limit of preoxidation was not carried out.

The other desired property of the preoxidized coals relates to the fluidization behavior. In order to operate the gasifier at a practical throughput and for the preoxidized coals to have a relatively high particle density, it is necessary to use a relatively coarse feed. These properties permit operation of the gasifier at reasonable gas velocities without excessive entrainment, and they maintain a reasonably high bed inventory to satisfy the demands of the gasification kinetics. It is seen from the data in Table 3 that significant particle swelling occurred in the preoxidation treatment of both coals.

Reduction in the amount of preoxidation required for Ireland Mine coal can be accomplished by use of finer coal. For the reasons cited above, however, the use of fine coal is economically undesirable.

The conclusion from these preoxidation studies was that Illinois coals may be pretreated successfully with pressure gasification by use of "adiabatic" preoxidation. Highly fluid Pittsburgh seam coals, however, require economically excessive amounts of preoxidation, unless an impractically small size consist feed coal is used.

Table 3. PROPERTIES OF COAL AND PREOXIDIZED COALS

Coal	Pittsburgh seam Ireland Mine coal		Illinois No. 6	
Treatment	Raw	28.3% Preoxidation at 750°F	Raw	8.7% Preoxidation at 810°F
% Sulfur	4.52	3.75	4.93	3.40
Ash	11.36	13.76	21.86	15.39[a]
Mean diameter, inch	0.0165	0.0156	0.0176	0.0175
Mean density, lb/ft³	81.0	56.4	80.0	51.8

[a] Ash is low because of segregation and selective removal of mineral matter in the preoxidizer.

Pretreatment via "Seeded Coal Process"

The principle of the preoxidation method of pretreatment is to convert the coal to a more rigid structure via oxidation, such that the fluidity is severely reduced when the coal undergoes pyrolysis.

The "Seeded Coal Process" would operate on just the reverse principle and actually utilize the natural fluidity of the coal. In the process visualized, char would be circulated at a high rate by means of a lift gas through a draft tube immersed in a normal fluidized bed. Coal and fine size seed char would be fed into the draft tube. The external fluid bed would be maintained at 1000 to 1400°F either by injection of air or hot fluidizing gas from a gasification step, as shown in Figure 1.

The coal melts, smears out over the surfaces of the seed char and external bed material, and then solidifies on completion of pyrolysis.

The demonstration of such a device[7] was successfully carried out in the low-temperature carbonization section of the CSF Coal Liquefaction Pilot Plant at Cresap, West Virginia. The feed material, in this instance, was somewhat different and constituted the underflow from the hydrocyclone separation of the extraction effluent. Coal extract in this case was used instead of the fluid coal; the extraction residue was used instead of the seed char. Other differences were that the mixture was sprayed into the draft tube as a slurry and operating temperatures and pressures were lower, i.e., 825 to 925°F and approximately 4 psig, respectively.

In this particular installation, a 36-in. ID carbonizer was employed in which there was installed a 6-in. ID x 11-ft high draft tube. Solids were circulated through the draft tube by injection of about 3500 to 4500 scfh of lift gas into the bottom of the tube. The feed was sprayed into the circulated char stream within the draft tube by means of a nozzle 3 feet above the lift gas injection point.

Solids circulation rates of the order of 100,000 lb/hr were achieved in this device, while complete operability and product size consist control was maintained with extract feed rates up to 200 lb/hr. The ratio of extraction residue solids to extract was in the range of about 1.5:1 to 3:1. The above throughput rates do not necessarily represent the capacity of the system since high extract feed rates were not available and consequently were not tested.

The above results led to an attempt to apply the same system to coal even though coal is a less fluid material than extract and the operating conditions, particularly the pressure, required are higher.

Tests with an inert bed of 48 x 100 mesh char at 1500°F and 15 atm system pressure showed that the external baffle was effective. The solids circulation rate upward through the tube was measured by substituting a known amount of air for some of the N_2 entering the solids feed line.[1C] From the measured temperature rise, the solids flow rate was calculated as 900 lb/hr by heat balance. Calculations involving the pneumatic transfer line model, devised in the course of development of the CO_2 acceptor project, showed that without the external baffle about 270 of the 340 scfh of N_2 fed to the bottom of the external bed had entered the draft tube; with the baffle, the flow was reduced to about 60 scfh.

Seven tests were made with the modified draft tube, using an external bed of 48 x 100 mesh char at 15 atm system pressure. Common conditions for the runs are listed below:

Ireland mine coal, sized to 100 x 200 mesh
Coal feed rate: 2.0 lb/hr
Duration of feeding: 3.3 minutes
Air to coal feed line equivalent to 100% of adiabatic preoxidation level at the temperature used.
Gas flows, scfh

Air + N_2 to coal feed line	65
N_2 to accelerating line	85
N_2 to bottom of external bed	340

Tests were made at temperatures from 900 to 1500°F, in 100 degree increments. Temperature limits of operability were established as follows: (1) at 900°F little or no smearing occurs as was shown by presence of coal-derived material in the form of hollow spheres in the bed after the run, and (2) at 1500°F caking occurred in the draft tube. Unfortunately, we were severely handicapped by the small scale of the equipment available, since the draft tube principle had to be adapted to the existing 4-in. diameter gasifier vessel.

The potential advantages of the process are that it will supply a feedstock that is assuredly operable with respect to agglomeration at gasifier conditions; and it can produce a dense, closely sized feedstock substantially free of fines. This will permit a higher gasifier throughput than otherwise.

A series of exploratory tests were carried out with the configurations A, B, and C indicated in Figure 3. Best results were obtained with configuration C, but even here two basic deficiencies were noted. From the appearance of the agglomerates obtained, it was apparent that insufficient mixing occurred in the draft tube between the injected coal and the circulating char. Part of the difficulty is associated with the small scale of the equipment, since calculations show that the Reynolds number in the draft tube is barely above the Stokes Law range. Also, it was apparent that most of the fluidizing gas bypassed the main bed in favor of the draft tube. The result was that a fluidized bed was not maintained external to the draft tube.

To overcome these limitations, the configuration C of Figure 3 was modified as follows:

To allow installation of an external baffle which would maintain fluidization of the external bed, the draft tube was raised 2 inches and the inlet lines were lengthened accordingly. An elliptical baffle 3-5/8 x 1-3/4 x 1/16-in. thick was welded to the accelerating gas line below the mouth of the tube at a slope of 60° from the horizontal. To help promote mixing a conical baffle was installed inside the tube with the apex of the cone positioned 1/2-in. above the end of the coal inlet tube.

The products from the runs at 1000 to 1400°F all showed more uniform smearing than in any of the previous runs without the internal baffle. At the end of each run, the system was depressured and the bed was drained by removing the coal inlet line. The hot bed material was quenched rapidly by contact with dry ice in the catchpot. The entire bed material then was screened at 28 and 48 mesh. All the run products contained some +48 mesh agglomerates which were external bed particles cemented together by a thin film of coal-derived-material. No agglomerates larger than 28 mesh were found. The fewest agglomerates occurred at 1300°F, indicating that this may be the optimum temperature with respect to uniformity of smearing. The amounts of +48 mesh agglomerates which formed are listed below:

Temperature, °F	+ 48 Mesh Agglomerates, wt % of Bed Inventory
1000	18.0
1100	16.0
1200	15.5
1300	8.0
1400	10.7

The particle density, measured in mercury, for the +48 mesh agglomerates formed at 1300°F had a high value of 85 lb/ft^3.

An attempt was made to run for a prolonged period at 1300°F and 15 atm system pressure to determine the size distribution of the "equilibrium" product. To simulate the seed char in the commercial embodiment (fines from the internal cyclones in the gasifier) an initial external bed of -100 mesh precarbonized char was established. Then, 100 x 200 mesh Ireland Mine coal and additional -100 mesh char were fed to the draft tube at rates of 2 and 4 lb/hr, respectively. The fine char contained a considerable amount of -325 mesh material which was elutriated from the reactor. The outlet piping system of the present equipment was not designed to handle large amounts of solids. The run had to be terminated after 35 minutes of feeding coal because the outlet system began to plug. Thus, an equilibrium bed was not established. However, analysis of the bed showed that it contained 50 weight percent of +100 mesh agglomerates, with a top size of 24 mesh.

The high particle density achieved is favorable, in that smearing of liquid coal over the seed particles apparently occurs as desired.

The small size of the existing equipment precludes any further meaningful studies of the seeded coal process. The radial clearance between the inlet line and the wall of the draft tube is only 0.15 inch. The mouth of the tube eventually would become choked by the larger agglomerates which inevitably would be formed.

The results of the exploratory studies strongly indicate that future studies should be made.

Several essential factors are required to achieve success in such an operation. Intensive mixing in the draft tube is required to achieve smearing of the "liquid" coal over both the seed and recirculating char. A sufficient residence time in one pass through the unit of the recirculating burden is needed to complete the "drying out" or carbonization of the coal. Finally, the draft tube must be large enough to handle the largest size particles made in the process without choking. All these factors point to a need for a larger unit in which the draft tube would be at least 2 inches in diameter as opposed to the present 0.680 inch. Such a unit, of course, would have a much higher capacity for coal feed which would lie approximately in the range of 30 to 300 lb/hr.

Pretreatment via Pre-Extraction

This method would be a more direct application of the draft tube pyrolysis method already demonstrated at the CSF pilot plant at Cresap, West Virginia. Two principal differences would be required here. First of all, the extraction slurry would be injected into the draft tube unit operated at 15-20 atm pressure instead of at substantial atmospheric pressure, and secondly, the ratio of extract to extraction residue normally would be greater since little or no extract need to be recovered as such. This technique would possibly prove to be more operable.

Reaction Kinetics

The other limiting factor is the gasification of bituminous coals via the CO_2 acceptor process is the poor reaction kinetics relative to sub-bituminous coals and lignites. The treatment of the gasification kinetics is described in more detail in a companion paper[8] presented at this conference which deals with gasification of bituminous coals to produce low-Btu gas. Suffice to say, that data available to us indicate that gasification rates with bituminous coal chars are about 1/15th of lignite chars. This necessitates increasing the gasification temperature to 1650°F to achieve adequate rates. There is little incentive from the kinetic point of view to increase the temperature with the available size and density of the char treated, i.e., 28 x 100 mesh and 45 lb/ft^3 particle density. The limiting factor in throughput in this instance becomes the fluid dynamics of the char particles rather than kinetics of gasification. The use of coarser feedstocks, of course, would remove this limitation and would require higher gasification temperatures to achieve higher outputs.

The acceptor process becomes deficient in heat supplied to the gasifier at this high temperature (1650°F) unless one or both of the following expedients is employed. More sensible heat as opposed to chemical heat may be supplied by increasing the acceptor circulation rate; or heat may be supplied by recycle of carbon dioxide.

The amount of supplementary heat required by either of the two above expedients also may be lowered by increase in operating pressure.

Process Description

An outline of the proposed process is given in Figure 1 previously mentioned. This incorporates the system of CO_2 recycle to supply the heat deficiency in the gasifier, and the "countercurrent" contacting of the feed coal with the gasifier offgas in the draft tube pretreater. This latter step not only "decakes" the coal feed but also significantly increases the Btu of the product gas.

The heat and material balance relationship is given in Table 4 for the processing of bituminous coal via the flow scheme of Figure 1. The heat and material balances were derived by adaptation of the computer program as previously devised for the OCR project on the development of the CO_2 acceptor process. The process assumptions used are generally quite similar to those outlined in the companion paper.[8]

Table 4. HEAT AND MATERIAL BALANCE IN TREATMENT

	Stream number							
	1	2	3	4	5	6	7	8
Product	Steam	CO	Recycle gas	Pre-treater char	Calcined acceptor	Gasifier except recycle	Spent acceptor	Fuel char
°F	1200	1200	400	1250	1873	1650	1650	1650
moles	2.759	1.100	0.157	4.222	5.750	5.064	0.230	2.124
lb				60.9				37.3
Composition,								
mole %								
CH_4			Same			3.01		
H_2			as			46.43		
CO			10			21.30		
CO_2						7.52		
H_2O						21.74		
H_2S						-		
N_2						-		
MgO·CaS					27.2		27.2	
$MgO \cdot CaCO_3$					-		25.5	
MgO·CaO					72.8		47.3	
Hydrogen				5.01				2.61
Carbon				94.99				97.39

[a] Dry, H_2S-free basis. HHV = 424 Btu/ft^3.

[b] Total sulfur content = 100 ppm.

[c] Water content only. Dust, tar, and phenols content not known.

System pressure: 14.84 atm (204 psig).

Basis: 100-lb dry Ireland Mine coal

	wt % (dry basis)	mole
H	4.8	2.381
C	69.8	5.812
N	1.2	-
O	7.6	0.475
S	4.3	0.1341
Ash	12.3	-

6 percent moisture, as fed; 50 percent of coal sulfur removed in preheater; 95 percent carbon burnout in regenerator; cold efficiency, 79.9 percent; total carbon gasified, 64.4 percent; and fixed carbon gasified, 56 percent.

OF BITUMINOUS COAL VIA CO_2 ACCEPTOR PROCESS (FIGURE 1)

Stream number								
9	10	11	12	13	14	15	16	17
Pretreater gas except recycle	Water gas	Air	Regenerator gas including lift	Absorber gas	Makeup acceptor	Dirty liquor	Lift gas	Excess gas
1250	100	350	1873	-	60	200	1200	-
7.120	6.279	9.051	13.548	4.538	0.230	0.841	2.500	0.938
					48.1			
17.15	19.63[a]		-	Same			-	
40.41	46.25		-	as			-	as
19.92	22.80		1.98	12			2.7	16
9.89	11.32		27.27				2.5	
11.70	-	0.44	0.78			100[c]	0.4	
0.93	-		b				-	
-	-		69.97				94.4	

The gasification temperature 1650°F is compatible with an overall gasification rate of: $R_T = 39 \times 10^{-4}$ lb fixed carbon gasified/lb carbon in bed/minute, and the other conditions cited with respect to carbon and steam conversions, etc.

Recent experiments under the EPA contract with the seeded coal process show that the pretreated coal from the draft tube (actually, char, since the temperature is in the range of 1200 to 1300°F) will have a high particle density of about 85 lb/ft^3. The particle density of the gasifier bed then would be about 45 lb/ft^3 after gasification of 56 percent of the fixed carbon (see Table 4).

Calculations involving the gas flow rates, the above gasification rate, and our fluidized-bed density correlation[1c] showed that one train with the gasifier bed dimensions shown below will be capable of processing 272,000 lb/hr of MF coal.

Fluidized bed height:	50 ft
Gasifier ID:	25.4 ft
Bed density:	14.7 lb/ft^3 char
Fluidizing velocity:	0.81 ft/sec

The estimated Btu content of the dry product gas is 424 Btu/ft^3 and the overall thermal efficiency on a cold gas basis is 79.7 percent. Thus, a single train is capable of producing about 1.5×10^8 ft^3 or 6.5×10^{10} Btu/day of raw gas. This compares favorably with the 3.5×10^{10} Btu/day (22.5-ft-ID gasifier) raw gas output projected for a single train in the CO_2 acceptor process when used for lignite gasification.

There are a few features incorporated in the process which should be mentioned.

A small amount of recycle gas (Stream 3) is added to the gasifier boot to prevent oxidation of CaS in the recirculating acceptor stream to $CaSO_4$ by the incoming steam and CO_2.

The fate of the coal sulfur is not known at present. It was assumed for the purposes of the balances that are presented that half of the sulfur is eliminated as H_2S in the pretreater; it will have to be removed by scrubbing the product gas.

The regenerator is operated sufficiently on the reducing side, such that the remaining sulfur is largely retained and discarded in the spent acceptor as CaS. It also will be recovered as H_2S via the Claus-Chance reaction as proposed previously.[1d]

The sulfur content in the regenerator off-gases is low enough, such that it may be flared without scrubbing; but prior incineration of residual reduced sulfur forms to SO_2 is required.

The system as shown here is certainly not optimum and some improvements are potentially possible as listed below:

1. Increase operating pressures from 18-20 atm. This will reduce the quantity of CO_2 recycle needed for heat balance and also raise Btu content of the product gas. As offsetting features, higher regenerator temperatures and a system to recycle product gas to the gasifier would be required.
2. The CO_2 recycle requirement could be derived from the product gas. Sufficient CO_2 would be removed from the product gas, after water-gas shift and prior to methanation to provide the CO_2 recycle requirements. As a matter of fact, the hot pot absorbent could be regenerated in such a way as to generate directly the steam-CO_2 mixture required for gasification under full system pressure, thus eliminating the CO_2 compressor. A difficulty here is the presence of H_2S in the product gas. Processes are available, however, which afford at least partial selectivity in H_2S versus CO_2 removal.
3. Recycle tar oil and tar to pretreater rather than the gasifier. It would be used as a vehicle to pressurize and transport the coal to the pretreater. By preheating the slurry to obtain partial extraction, improved operability may be achieved.

4. By use of a catalytic afterburner to combust the CO in the regenerator offgas, it may be possible to generate surplus power through the expansion turbine. An afterburner will be required in any case to convert traces of H_2S, COS, and S_2 in this gas to SO_2.

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BIBLIOGRAPHY

1. Colsolidation Coal Co., Research & Development Report No. 16 to the Office of Coal Research, U.S. Dept. of the Interior, Washington, D.C. under Contract Number 14-01-0001-415.
   a. Interim Report No. 1. Pipeline Gas from Lignite Gasification — A Feasibility Study. U.S. Dept. of Commerce, National Technical Information Service, PB-166817 (feasibility study), PB-166818 (appendix). February 1965.
   b. Interim Report No. 2. Low-Sulfur Boiler Fuel Using the Consol CO Acceptor Process. U.S. Dept. of Commerce, National Technical Information Service PB-176910. November 1967.
   c. Interim Report No. 3. Phase II — Bench-Scale Research on CSG Process. January 1970.
      Book I, Studies on Mechanics of Fluo-Solids Systems. U.S. Government Printing Office Catalog Number 163.10:16/INT3/Book 1.
      Book 2, Laboratory Physioco-Chemical Studies. U.S. Government Printing Office Catalog Number 163.10:16/INT3/Book 2.
      Book 3, Operation of the Bench-Scale Continuous Gasification Unit. U.S. Government Printing Office Catalog Number 163.10:16/INT3/Book 3.
   d. Interim Report No. 4. Pipeline Gas from Lignite Gasification — Current Commercial Economics. U.S. Government Printing Office Catalog Number 164.10:16/INT4.
2. Curran, G.P., J.T. Clancey, C.E. Fink, B. Pasek, M. Pell, and E. Gorin. Annual Report to Office of Air Programs, Environmental Protection Agency under Contract Number EHSD-71-15. September 1, 1970 to November 1, 1971.
3. Curan, G.P., W.E. Clark, and E. Gorin. Low-Sulfur Char as a Co-Product in Coal Gasification. Environmental Protection Agency, Research Triangle Park, N.C. EPA-R2-76-060, October 1972.
4. Curran, G.P., C.E. Fink, and E. Gorin. Proceedings of Second International Conference on Fluidized-Bed Combustion, 1970. Office of Air Programs, Environmental Protection Agency, Research Triangle Park, N.C. Publication Number AP-109. pp. III-1-1 to III-1-11.
5. Unpublished work carried out jointly by Consolidation Coal and Esso Research.
6. Forney, A.J., S.J. Gasior, R.F. Kenny, and W.P. Haynes. Proceedings of the Second International Conference of Fluidized-Bed Combustion, October 1970. Office of Air Programs, Environmental Protection Agency, Research Triangle Park, N.C. Publication Number AP-109. pp. III-3-1 to III-3-21.
7. Pilot scale Development of the CSF Process. Period July 1, 1968 - December 31, 1970, R & D Report No. 39, Volume IV, Book 3. Consolidation Coal Co. Prepared for Office of Coal Research, U.S. Dept. of the Interior, Washington, D.C. under Contract Number 14-01-0001-310.
8. Curran, G.P., B. Pasek, M. Pell, and E. Gorin. Low-Sulfur Producer Gas via an

Improved Fluid Bed Gasification Process. (Presented at 3rd International Conference on Fluidized-bed Combustion, October 1972.) (Session III, Paper 6, this document.)

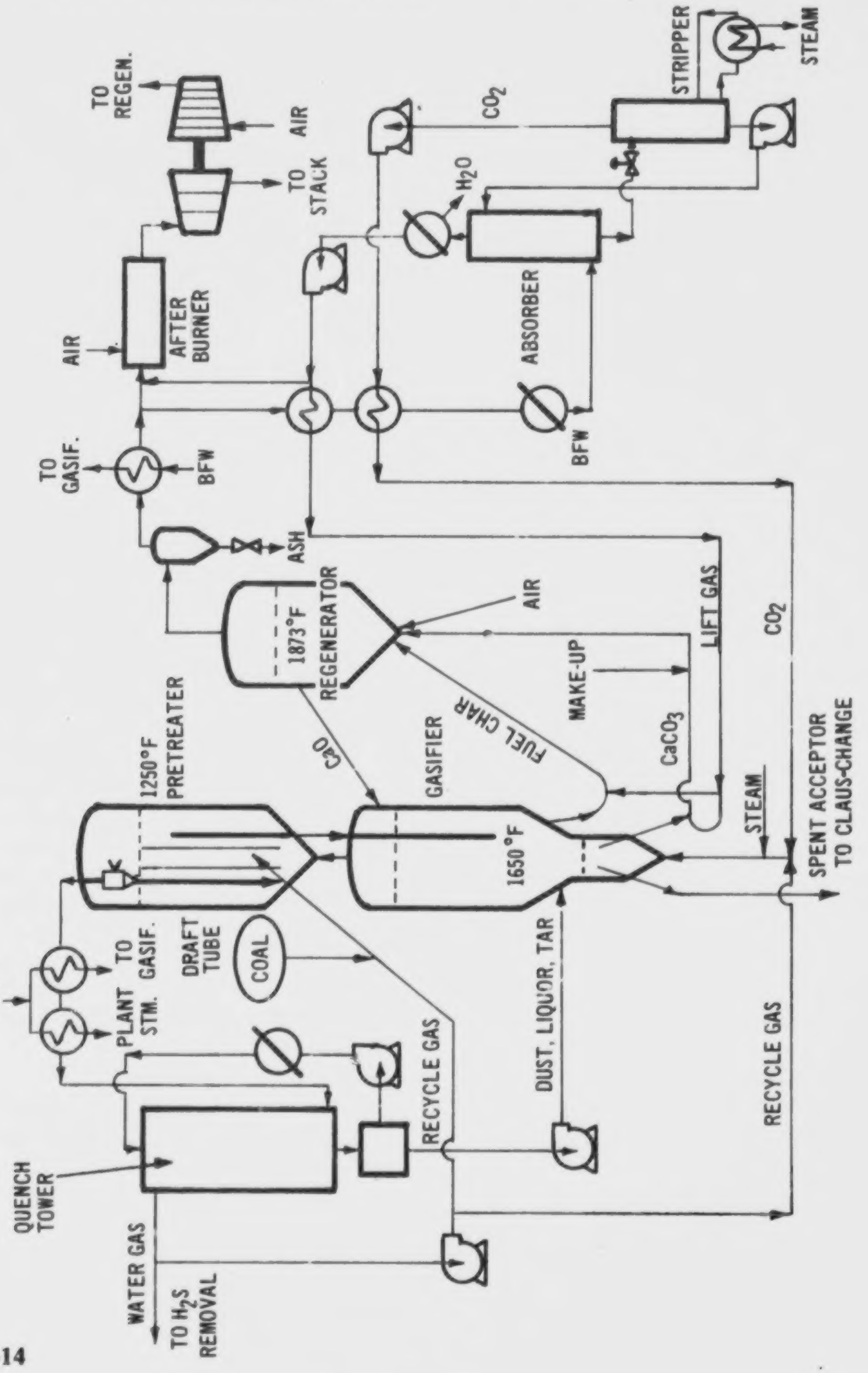


Figure 1. Processing of bituminous coal by the CO_2 acceptor process.

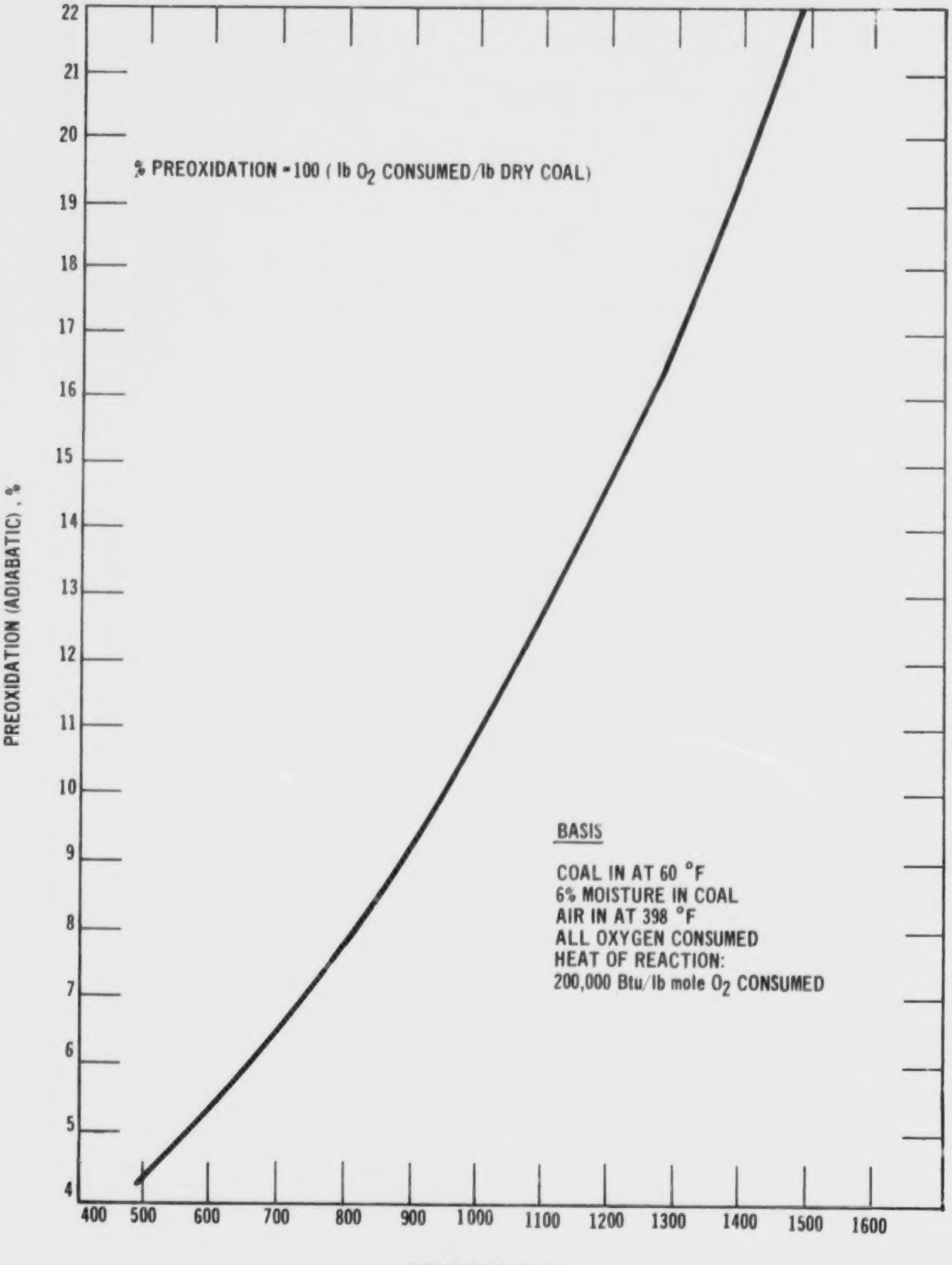
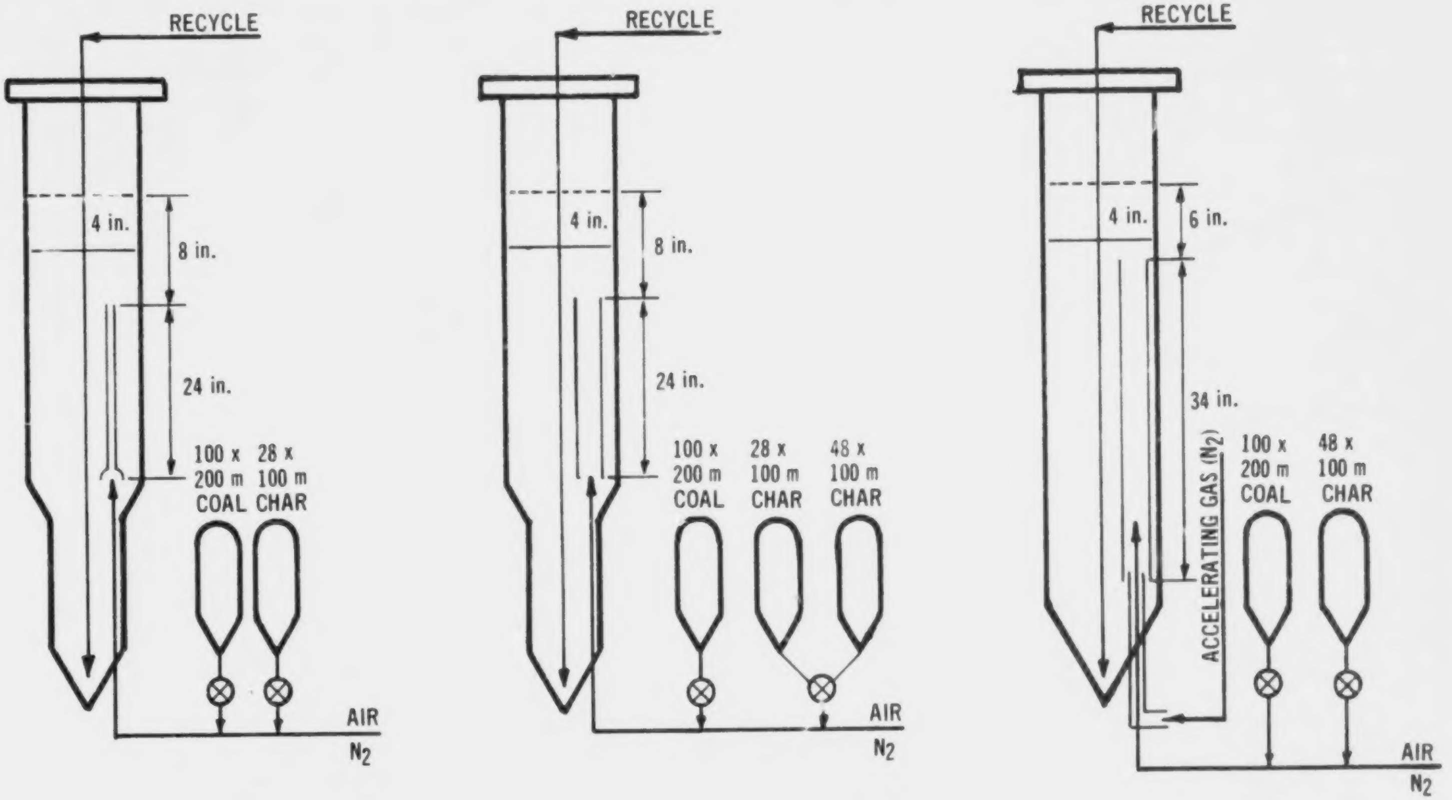


Figure 2. Percent preoxidation versus temperature for adiabatic constraint.



	CONFIGURATION A	CONFIGURATION B	CONFIGURATION C
DRAFT TUBE	0.500 in. OD x 0.444 in. ID	0.75 in. OD x 0.680 in. ID	0.750 in. OD x 0.680 in. ID
COAL FEED LINE	0.250 in. OD x 0.180 in. ID	0.250 in OD x 0.180 in. ID	0.250 in. OD x 0.180 in. ID
TIP POSITION	HALFWAY INTO SKIRT	1 in. ABOVE TUBE BOTTOM	5 in. ABOVE TUBE BOTTOM
ACCELERATING GAS LINE			0.375 in. OD x 0.305 in. ID (TIP POSITION 1 in. ABOVE TUBE BOTTOM)

Figure 3. Configuration of draft tubes used in seeded coal tests.

6. LOW-SULFUR PRODUCER GAS VIA AN IMPROVED FLUID-BED GASIFICATION PROCESS

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ABSTRACT

This paper describes the evolution of the process concepts for generation of clean low-Btu gas from bituminous coals via fluid-bed gasification. The improved process now under development for the EPA does not involve the CO_2 acceptor principle. Hot sulfur recovery from the gas is achieved by the use of dolomite. The residual char from the gasifier is utilized in a carbon burn-up cell. The heat sink utilized in this case is the sensible heat of the air and steam feed to the gasifier.

Dolomites show activity for hot sulfur cleanup via the reaction,

$$CaCO_3 + H_2S = CaS + H_2O + CO_2.$$

A single limestone was tested and was substantially inactive.

Various dolomites have been assessed and best results are obtained with pure crystalline type stones.

Experimental background data around other key process steps are also briefly presented.

INTRODUCTION

The production of low-sulfur producer gas via an adaptation of the CO_2 acceptor process was described in a paper[1] given at the Second International Conference on Fluidized-Bed Combustion. The system was described in some detail along with some supporting background experimental data.

A detailed process design and feasibility study of the sytem as well as an experimental evaluation was carried out under a contract with the EPA. The results are reported in detail in the Annual Report.[2] The economics will be briefly summarized later in this report.

The experimental evaluation of the system indicated feasibility of all steps in the process with one exception. It was found that the sulfur recovery from the acceptor would be incomplete from the regenerator. This necessitated addition of another step in the process in which sulfur is rejected by the reaction first proposed by Squires,[3]

$$CaS + H_2O + CO_2 = CaCO_3 + H_2S. \qquad (1)$$

With this added complication introduced, further thought was given to refining and simplifying the overall process.

It became apparent that there is no real advantage in using the CO_2 acceptor reaction simultaneously with the sulfur acceptor reaction in the gasifier when low-Btu fuel gas is the desired product. Disposition of the residual char from the gasifier can be accomplished by use of a carbon burn-up cell which preheats all the steam and air required for the gasifier. The sensible heat duty involved in preheating serves as the "heat sink", which is necessary to prevent ash slagging during combustion of the residual gasifier char. In the CO_2 acceptor process the heat sink is provided by the endothermic calcining reaction in the acceptor regenerator.

Since the acceptor no longer needs to be in the gasifier, the sulfur acceptance reaction now can be carried out in a separate, external reactor containing a dense-phase fluidized bed of dolomite in the form of $CaCO_3 \cdot MgO$.

IMPROVED FLUID-BED PROCESS DESCRIPTION

A schematic diagram of the revised process is shown in Figure 1. The simplest configuration occurs when the burn-up cell is integral with the gasifier. In this instance, it would be analogous to the CO_2 acceptor gasifier in that the burn-up cell would be in the form of a "boot" which would contain a fluid bed of coarse inert solids such as "dead burned lime." Combustion of the char residue from the gasifier would take place in the boot. In figure 1, the burn-up cell is shown as a separate reactor. This is a costlier configuration but permits more selective rejection of ash. The hot fuel gas is desulfurized in the H_2S sorption bed by the reaction,

$$H_2S + CaCO_3 = CaS + H_2O + CO_2. \quad (2)$$

The bed temperature is held at a level below which the acceptor can calcine by the reaction,

$$CaCO_3 = CaO + CO_2. \quad (3)$$

The low-sulfur hot gas is cooled to 1300°F by heat exchange with the water needed to generate the gasifier steam, and then is cleaned of particulates and alkali by high pressure drop cyclones.

The sulfided acceptor is conveyed to the regenerator by continuously recirculating a stream of CO_2 and steam. In the regenerator the "Squires" reaction takes place at about 1300°F,

$$CaS + CO_2 + H_2O = CaCO_3 + H_2S. \quad (4)$$

The regenerated acceptor is returned by gravity to the sorption reactor.

A computer program has been devised to evaluate the heat and material balance relationships and overall thermal efficiency of the scheme shown in Figure 1.

The program evaluates the interaction between the various components of the system consistent with the thermodynamic, fluo-solids mechanic and kinetic restraints on the system.

The entire system was represented by 27 simultaneous linear and non-linear equations which represent the five basic process steps given below:

1. Carbon burn-up cell,
2. Gasifier,
3. Sulfur reactor,
4. Steam-product gas exchanger,
5. Squires reaction - impact of temperature only,

which also are interrelated by the following quantities:

1. H, C, and O balance,
2. Heat balances around components 1 through 4, above,
3. Water-gas-shift equilibrium in components 2 and 3, above,
4. Methane yield correlation,
5. Equilibrium in the reaction,

$$CaCO_3 + H_2S = CaS + CO_2 + H_2O. \quad (5)$$

The above equations were solved by an iterative procedure for the moles of air and steam fed to the burn-up cell as a function of the following variables:

1. Burn-up cell temperature.
2. Gasifier temperature.
3. S/Σ Ca, mole ratio.
4. "Squires" reactor temperature.

Once having computed the input and output flows and compositions by the method outlined above, it was necessary to determine the gasification reactor sizes. The vessel sizing is determined by the interaction of the fluidization mechanics of the char particles and the gasification kinetics.

The basis used here was to provide for a single train to process 120,800 lb/hr of coal. The fluidized-bed height was fixed at 50 feet. The fluidized-bed density was then calculated using the correlation developed during the work on the CO_2 acceptor process. It was assumed here that a high-density, closely sized char particle would be generated by the "seeded coal process" from Ireland Mine coal (the mean particle diameter was taken as 0.04

inch and the initial particle density of 85 lb/ft^3). The reduction in particle density as affected by carbon burnoff was based on the relationships developed during the work on the CO_2 acceptor process.[6]

It is now necessary to compute the vessel cross section and fluidizing velocities which are compatible with the estimated bed inventories by use of reaction kinetic data.

Extensive differential rate data[7-9] were obtained some time ago on the gasification of bituminous coal chars as a function of temperature, pressure and mole fraction of hydrogen in hydrogen-steam mixtures. Subsequently, extensive kinetic data were obtained on the gasification kinetics of lignite char.[6,10] In this work it was found that the reaction rate was strongly inhibited by the presence of CO as well as hydrogen. Thus, the prior data on bituminous coal chars could not be used since the inhibiting effect of CO was not taken into account. However, under comparable conditions (in the absence of CO) it was found that on the average the bituminous coal chars had about 1/15th the reactivity of the lignite chars.

Therefore, in developing the kinetic calculations the equations developed for lignite char[10] were used with introduction of a correction factor of 1/15th to account for the lower reactivity of the bituminous coal chars. The differential kinetics were translated into integral kinetics; i.e., they were averaged over the whole bed by the method given in Appendix D of the Annual Report.[2]

Having calculated the integral rate R_T, the required bed inventory is calculated from the equation,

$$R_T = \frac{\text{lb fixed carbon gasified}}{\text{min/lb fixed carbon in bed}} \times 10^{-4}$$

The fluidization calculations outlined above are then used to determine the required vessel cross section and fluidizing velocity.

A total of 14 cases were computed covering gasification temperatures between 1650 and 1750°F. Because of the kinetic and thermodynamic limits the system is highly constrained, and the response of the system is quite limited. This is illustrated by the ranges given in Table 1.

Table 1. RANGE OF VARIABLES STUDIES IN SYSTEM ANALYSIS

Range of independent variables studied	Base Case	
Gasifier temperature, °F	1650-1750	1725
Burn up cell temperature, °F	1750-1950	1883
S/Σ Ca, mole ratio	0.1-0.4	0.2
Squires reactor temperature, °F	1200-1300	1300
Range of calculated quantities		
Cold gas efficiency, %	79.1-81.2	80.6
HHV dry product gas, Btu/ft^3	143-149	147
Sulfur removed, %	93.1-97.5	96.7
Steam conversion, %	49.4-55.7	53.6
Carbon to burn up cell, wt % of C in coal feed	14.3-16.9	15.4
Gasification rate $\frac{\text{lb C (gasified)} \times 10^4}{\text{lb C in bed, min}}$	45-78	68
Gasifier, ID, ft	24.9-25.3	25.1
Char particle density	28.7-35.8	30.2
Gas fludizing velocity, ft/sec	1.46-1.52	1.49
Gasifier cross section index[a]	392-415	398
Constant parameters		
System pressure, atm	← 15 →	
Gas outlet temperature, °F	← 1300 →	

[a] Ft^2/10^9 Btu/hr (HHV of product gas).

The conditions for the base case, which is felt to be close to a practical "optimum" for the system, are also given in Table 1. A more complete heat and material balance around the base case is also given in Table 2.

The process concept given here has several potential advantages over the original process which utilized the CO_2 acceptor process, as outlined below:

Operability May Be Improved in the New Process

1. The O_2 partial pressure to the burn-up cell is lower than to the previous regenerator. Steam and air N_2 serve as the heat sink. There is less chance of ash slagging, especially since the burn-up cell can be operated at much lower temperatures than are needed to regenerate $CaCO_3$.

Table 2. KEY STREAM FLOWS AND ANALYSES, CASE 6

	Stream Number										
	1	2	3	4	5	6	7	8	9	10	11
Identification	Air	Steam	Fuel char	Burn-up cell gas	Raw product gas	Spent acceptor	"Squires" offgas	Regenerated acceptor	Sulfur	CO_2	Clean product gas
^oF	398	870	1725	1883	1725	1624	1300	1300	320	200	1300
moles	11.41	3.504	0.936	14.68	20.37	0.648	3.522	0.648	0.1296	0.1296	20.50
lb			23.14						4.15		
Composition, mole %											
CH_4				×	1.72		×				1.71
H_2				×	16.31		×				16.56
CO				×	18.96		×				18.50
CO_2				6.10	7.85		48.16				8.78
H_2O	0.40			24.42	10.06		48.16				10.29
H_2S				×	0.66		3.68				0.02
N_2				61.31	44.18		×				43.90
O_2				8.17	×		×				×
NH_3					0.26						0.25
$MgO \cdot CaS$						20		0			
$MgO \cdot CaCO_3$						80		100			
Hydrogen (as H_2)			4.25								
Carbon			95.76								

Basis: Ireland Mine Coal (100 lb dry coal)

	wt %, dry basis	moles
H	4.8	2.381
C	69.8	5.812
N	1.2	-
O	7.6	0.475
S	4.3	0.1341
Ash	12.3	-

6% moisture as fed; system pressure: 15 atm (206 psig)

2. The O_2 partial pressure to the gasifier is lower, since the air is diluted with all of the input steam and all of the products of combustion of the burn-up cell. Thus, one possibly can raise the temperature to 1750°F to improve kinetics without increasing the danger of ash slagging.

3. More positive contact of dirty gas with the dense-phase bed of acceptor in sulfur reactor is effected. Also, in certain circumstances it may be possible to use the sulfur reactor as a fluid-bed filter to remove "residual" particulate matter.

The Cold Gas Efficiency is Definitely Improved

1. Lower duty to calcine make-up acceptor. Circulation rate is about 10 percent that of the original concept.
2. 100 percent burn up of carbon (versus 98 percent).
3. Improved gasification kinetics require less steam. Thus, less latent heat in product gas.
4. Less air required. Thus, less sensible heat is lost with N_2.

EXPERIMENTAL BASIS

Pretreatment

This was discussed in the preceding paper[4] and only the conclusions need be reiterated here. Pretreatment by preoxidation is a viable procedure for the more weakly caking Illinois No. 6 coals, but is not a desirable procedure for use with the more highly fluid Pittsburgh Seam coals. For the latter coals, pretreatment by the seeded coal process appears promising but further development to prove out the method is required.

Gasification Operability and Kinetics

Studies were carried out to demonstrate operability of the gasifier with respect to both caking and ash fusion using pretreated Illinois No. 6 coals. The conditions studied were those that correspond to the original adaptation of the CO_2 acceptor process to low-Btu gas production. Complete operability in the 4-in. diameter gasifier was achieved on both points.

It is felt that the conditions in the present system are less stringent, as was pointed out above, such that operability problems due to ash fusion are less likely to occur.

The background data on differential kinetics which were used to calculate integral gasification rates were outlined in the previous section. Integral gasification rates were obtained also in operation of the continuous unit with both Disco char and Illinois No. 6 coal.[4] The results were given in the Annual Report.[2] The results are in approximate agreement with the basis used for reactor design given above, although the rates obtained with Disco char tend to be somewhat lower than those obtained with the pretreated Illinois coal. Further data on the kinetics of gasification of bituminous coals are required to provide a firmer basis for reactor design.

Carbon Burn-up Cell

No data on the operability of this unit are available at this time although it is planned to obtain such data in the course of the present EPA contract. The operation comprises combustion at full system pressure of residue char in the presence of a fluid bed of inert solids, such as "dead burned lime." An analogous operation is the regeneration of the CO_2 acceptor process where residue char is used as fuel for acceptor regeneration. Operability of this process has been demonstrated in prepilot scale work on the process. A full-scale pilot test, of course, is scheduled at the Rapid City pilot plant.

Desulfurization and Sulfur Recovery Steps

General

An experimental program to test these steps is now under way in our library bench-scale unit as specified in our present EPA contract.

Preliminary results are now available for desulfurization of simulated producer gas at about 1600°F by means of half calcined dolomite and for regeneration at 1300°F by means of the "Squires" reaction. These will be discussed below.

The offgas from the Squires reaction at the specified operating conditions (1300°F) is relatively low in H_2S content due to equilibrium limitations. Special techniques are required to recover sulfur from this gas economically without condensation of steam or removal of carbon dioxide. For this purpose, a liquid-phase Claus reaction was proposed using hot water under pressure as the reaction medium. This is the so-called "Wackenroder" reaction, and the system is described in detail in the Annual Report.[2]

Laboratory equipment to test this process is now being assembled but no results are as yet available to report.

Description of Experimental System

A flow diagram of the new experimental unit is shown in Figure 2. Acceptor in the form of $CaCO_3 \cdot MgO$ is fed continuously at a known rate to the top of the H_2S-sorption reactor via a pneumatic lift line. The carrier gas is recycled product gas and it does not pass through the fluidized bed in the sorption reactor. Hot, H_2S-laden, producer gas is fed to the bottom of the bed. Steam, N_2, CO_2, H_2, and H_2S are added to a stream of recycle gas to simulated the partial pressures of the various components of the product gas from the gasifier shown in the process flow diagram in Figure 1. The reactor, previously used as the CO_2 acceptor regenerator, is 3-in. ID with a bed height of 18 inches.

The sulfided acceptor is fed by gravity to the top of the Squires reactor and is regenerated while being fluidized in a stream of steam-CO_2. The reactor, previously used as the gasifier vessel, has been necked down from 4-in. to 2-in. ID and has a bed height of 48 inches.

Continuity of acceptor recirculation is maintained by withdrawal and feeding through parallel lockhoppers as shown in Figure 2. In both reactors the acceptor is fed to the top of the bed and is withdrawn from the bottom. The height of each fluidized bed is held at the desired level by means of a ΔP cell placed across the upper part of the bed, which actuates a solids control valve located below the acceptor standleg.

The product gas from either reactor can be monitored continuously for H_2S content by means of a dualprange infrared analyzer. The continuously recirculating inventory of acceptor is sampled periodically from both reactors and analyzed for CaS and $CaCO_3$.

Note that the physical arrangement of the two reactors is reversed from that shown in Figure 1, for the process reactors. From an experimental standpoint, it is immaterial which reactor is the upper vessel. It is possible that regeneration of the acceptor by the Squires reaction will require a greater acceptor retention time than that in the H_2S-sorption reactor. The existing pressure shell and electrical furnace for the lower reactor is considerably larger than that for the upper reactor. It was chosen to house the Squires reactor because the bed volume can be easily increased by a factor of 3 over that of the present design if initial operations show the need.

Preliminary Experimental Results

A series of experiments were carried out, all with conditions similar to those listed in Tables 3 and 4 for the gas desulfurizer and regenerator, respectively.

The first series of runs were conducted with Tymochtee dolomite which was used in the previous work on the CO_2 acceptor process.[6] A fundamental difficulty found here is the very high attrition rate, which ran as high as 18 percent of the acceptor fed per pass.

Run A4 (Tables 3 and 4) was made with air injection into the regenerator in an attempt to harden the stone by partial oxidation to

Table 3. GAS DESULFURIZER

	Run number	
	A4	A7
Acceptor	35 x 48 mesh Tymochtee dolomite	28 x 35 mesh Canaan dolomite
Feed rate, lb/hr (half calcined basis)	5.9	6.5
Solids residence time, min	32	33
Input, scfh		
Recycle to bed	130	175
H_2S	3.5	3.5
CO_2	35	54
H_2	60	73
H_2O	–	–
N_2	65	96
Purges (CO_2) to bed	5	5
Purges (N_2) above bed	15	15
Recycle, acceptor lift gas, above bed	92	71
Output, scfh in cycle	1-3	1-2
Exit gas rate, scfh (dry basis)	148	215
Composition, mole %		
H_2	18.3	17
CO	17.3	18
CO_2	12.1	12
N_2 (by difference)	52.1	53
H_2S	0.09	0.05
Outlet gas, top of bed composition, mole %		
H_2O	10.9	9.7
H_2	16.3	15.4
CO	15.4	15.9
CO_2	10.8	11.2
N_2	46.5	47.7
H_2S	0.08	0.04
Flow rate at top of bed, scfh	275	416
Fluidizing velocity, ft/sec	0.40	0.60
Attrition, % of feed rate	5.6	0.7
Duration of circulation with H_2S feed, hr	7.1	25.2
Removal of feed sulfur, %	97	97
% H_2S in outlet/equilibrium % H_2S	2.3	1.4
Conversion of acceptor/pass, mole %	23	19
Temperature = 1600°F, Pressure = 206 psig		

Table 4. REGENERATOR

	Run number	
	A4	A7
Solids residence time, min	38	37
Input, scfh		
H_2O	89	110
CO_2	82	110
Air	7.5	–
H_2	–	–
Purges (N_2) to bed	10	8
Purges (N_2) above bed	10	10
Purges (CO_2) above bed	5	5
Output, scfh in cycle	1-3	1-2
Exit gas rate, scfh (dry basis)	110	133
Composition, mole %		
CO_2	76.1	85.7
N_2	23.0	13.5
H_2S	0.8	0.7
S_2	0.05	–
Outlet gas, top of bed		
Composition, mole %		
H_2O	43.5	48.3
CO_2	47.3	47.8
N_2	8.6	3.5
H_2S	0.5	0.4
S_2	0.03	–
Flow rate, top of bed, scfh	186	228
Fluidizing velocity, ft/sec	0.53	0.64
Regeneration of acceptor, mole %	6.7	5.1
Temperature = 1300°F, Pressure = 206 psig		

$CaSO_4$ in situ. Prior work in the CO_2 acceptor process development showed that hardening of the stone occurs when this is done at higher temperatures due to the formation of a transient liquid in the CaS-$CaSO_4$ system.[6] The attrition rate apparently was somewhat reduced over the comparable run where air injection was not used but was still unacceptably high. The operating temperature (1300°F) was apparently too low to achieve hardening by the transient liquid mechanism.

A Tymochtee dolomite which had been hardened by cycling through the CO_2 acceptor process was also tested. This material showed the expected good resistance to attrition. Only preliminary results are available at this writing, but the activity of the stone appears to be less than that of the fresh Tymochtee dolomite.

A Nebraska limestone was also tested; it showed very low absorption of H_2S as compared with half calcined dolomites. This is in accord with prior laboratory studies by Ruth, et al. Other limestones such as BCR-1692 used in the Esso, Ltd. work will be tested to determine whether the low activity is generic to all limestones.

A pure dolomite from Canaan, Connecticut, was also tested under the conditions indicated in Table 3 and 4. This material showed excellent attrition resistance with an average weight loss of only 0.8 weight percent per pass through the system.

Both the Tymochtee and Canaan dolomites showed excellent fresh activity for removal of H_2S in the gas desulfurizer. Ninety-seven percent removal of H_2S was achieved in both cases. The removal of CaS in the regenerator was highly incomplete in both cases. Thus, it would appear that the kinetics of the Squires reaction is a limiting factor in this system and this factor requires more study.

The definition of results in terms of number of cycles is difficult because of the semi-continuous nature of the acceptor circulation, loss of material due to attrition, and sampling of material for analyses. An approximate, and "conservative" method for calculating the number of cycles was used. Each pass of a charge through the unit is calculated as a fractional cycle Δn using the relationship: Δn equals the actual amount fed divided by the internal inventory plus the actual external inventory.

The number of cycles that could be achieved with the Tymochtee dolomite were limited due to the high attrition losses. In the case of the Canaan dolomite, attrition was not the determining factor; the run was terminated when breakthrough of H_2S occurred in the gas desulfurizer.

The composition of the offgases from the gas desulfurizer and regenerator as a function of the number of cycles for runs A4 and A7 are given in Figures 3 and 4, respectively. In both cases the H_2S content of the regenerator offgases increases with the number of cycles. This, as we will show below, is due to accumulation of CaS on the acceptor and points to poor kinetics in the Squires reactor. The H_2S content of the desulfurizer offgas remains relatively steady until the acceptor becomes heavily loaded with sulfur towards the end of the run.

The sulfur content of the stones for the two Runs A4 and A7 as a function of cycle number are given in Figures 5 and 6, respectively. It is immediately clear that in both cases there is rapid buildup of CaS on the acceptor due to incomplete regeneration of the CaS. The efficiency of sulfur removal and recovery is further illustrated by the tabular data presented for Run A7 with Canaan dolomite in Table 5.

At 19 percent conversion per pass to CaS (Table 3, Run A7), the known percent of sulfur rejected (Table 5) makes possible a rough estimate of the Ca/S ratio required if fresh dolomite feed were added continuously to the system operated under the above conditions. The Ca/S ratio would be approximately 0.45.

Table 5. RUN A7 H_2S CONTENT OF EXIT GASES (DRY BASIS)

CANAAN DOLOMITE					
Gas desulfurizer			**Regenerator**		
Cycle No.	**H_2S, mole %**	**H_2S Removal, %**	**Cycle No.**	**H_2S, mole %**	**Recovery of H_2S Feed, %**
0.3	0.046	97	0.5	0.671	25
1.4	0.049	97	1.5	0.927	35
5.5	0.048	97	5.7	1.39	53
7.5	0.058	96	7.8	1.69	64[a]
8.6	0.064	96	9.2	1.40	53[a]
12.6	0.672	59	12.5	1.90	72[a]

[a] The condensate contained about 3% of the feed sulfur as elemental sulfur.

The acceptor at the end of Run A7 was nearly completely converted to CaS, hence the break-through observed in the desulfurizer. At the end of the run, 2 hours additional residence time was given to the batch of acceptor remaining in the regenerator. The CaS content of the acceptor was reduced from 85 to 76 mole percent. This indicates again that the poor kinetics in the Squires reactor is controlling. Better results should be achieved if longer residence times in the regenerator are used. It does appear that a considerable amount of inactive CaS is inevitably formed; at the present time, however, we are unable to clearly distinguish between CaS of low reactivity and the "dead" material.

It thus appears that high purity crystalline dolomites have acceptable physical strength and activity for use in the process. Economically acceptable make-up rates can be achieved at the proper operating conditions. The geographical distribution of dolomites with acceptable strength and activity is now being studied. A variable study is also planned with selected dolomites to determine optimum conditions for their use.

ECONOMICS OF PROCESS

No economic figures are available for the improved process. The potential economics of the CO_2 acceptor based process were given in the Annual Report.[2] Figure 7 is reproduced from that report. It gives the cost of low-Btu gas as a function of coal cost delivered to a 1200-MW boiler from a large-scale gasification plant. The figures are based on 1976 operation, 15 percent capital charges and 7.5 percent/yr escalation on materials and labor, and 7.5 percent/yr interest during construction and an operating factor of 70 percent.

The economics of the new process are expected to be somewhat better.

ACKNOWLEDGMENT

Appreciation is expressed to the Environmental Protection Agency for financial support of the work presented in this paper and for permission to publish the results given.

BIBLIOGRAPHY

1. Curran, G.P., C.E. Fink, and E. Gorin. Proceedings of Second International Conference on Fluidized-Bed Combustion, 1970. Office of Air Programs, Environmental Protection Agency, Research Triangle Park, N.C. Publication Number AP-109. pp. III-1-1 to III-1-11.

2. Curran G.P., J.T. Clancey, C.E. Fink, B. Pasek, M. Pell, and E. Gorin. Annual Report to Control Systems Division, Office of Air Programs, Environmental Protection Agency, Research Triangle Park, N.C., under Contract Number EHSD-71-15. September 1, 1970-November 1, 1971.

3. Squires, A.M. Fuel Gasification. Advances in Chemistry Series No. 69. American Chemical Society. Washington, D.C., 1967. pp. 205-229.

4. Curran, G.P. and Everett Gorin. The CO_2 Acceptor Process — A Status Report. (Presented at 3rd International Conference on Fluidized-Bed Combustion. Hueston Woods. October 29-November 1, 1972.) See Session 3, Paper 5 this volume.

5. Book 1, Studies on Mechanics of Fluo-Solids Systems. Consolidation Coal Co., Research & Development Report No. 16 to the Office of Coal Research, U.S. Dept. of the Interior, Washington, D.C., under Contract Number 14-01-0001-415. Government Printing Office Catalog Number 163.10:16/INT3/Book 1.

6. Book 3, Operation of the Bench-Scale Continuous Gasification Unit. Consolidation Coal Co., Research & Development Report No. 16 to the Office of Coal Research, U.S. Dept. of the Interior, Washington, D.C., under Contract Number 14-01-0001-415. Government Printing Office Catalog Number 163.10:16/INT3/-Book 3.

7. Goring, G.E., G.P. Curran, R.P. Tarbox, and E. Gorin. Ind. Eng. Chem. 44:1051, 1057, 1952.

8. Goring, G.E., G.P. Curran, C.W. Zielke, and E. Gorin, Ind. Eng. Chem. 45:2586, 1953.

9. Zielke, C.W. and E. Gorin. Ind. Eng. Chem. 47:820, 1955, and 49:396, 1957.

10. Book 2, Laboratory Physico-Chemical Studies. Consolidation Coal Co., Research & Development Report No. 16 to the Office of Coal Research, U.S. Dept. of the Interior, Washington, D.C. Government Printing Office Catalog Number 163.10:16/INT3/Book 2.

11. Ruth, L., A.M. Squires, and R.A. Graft. Environmental Science and Technology. November 1972.

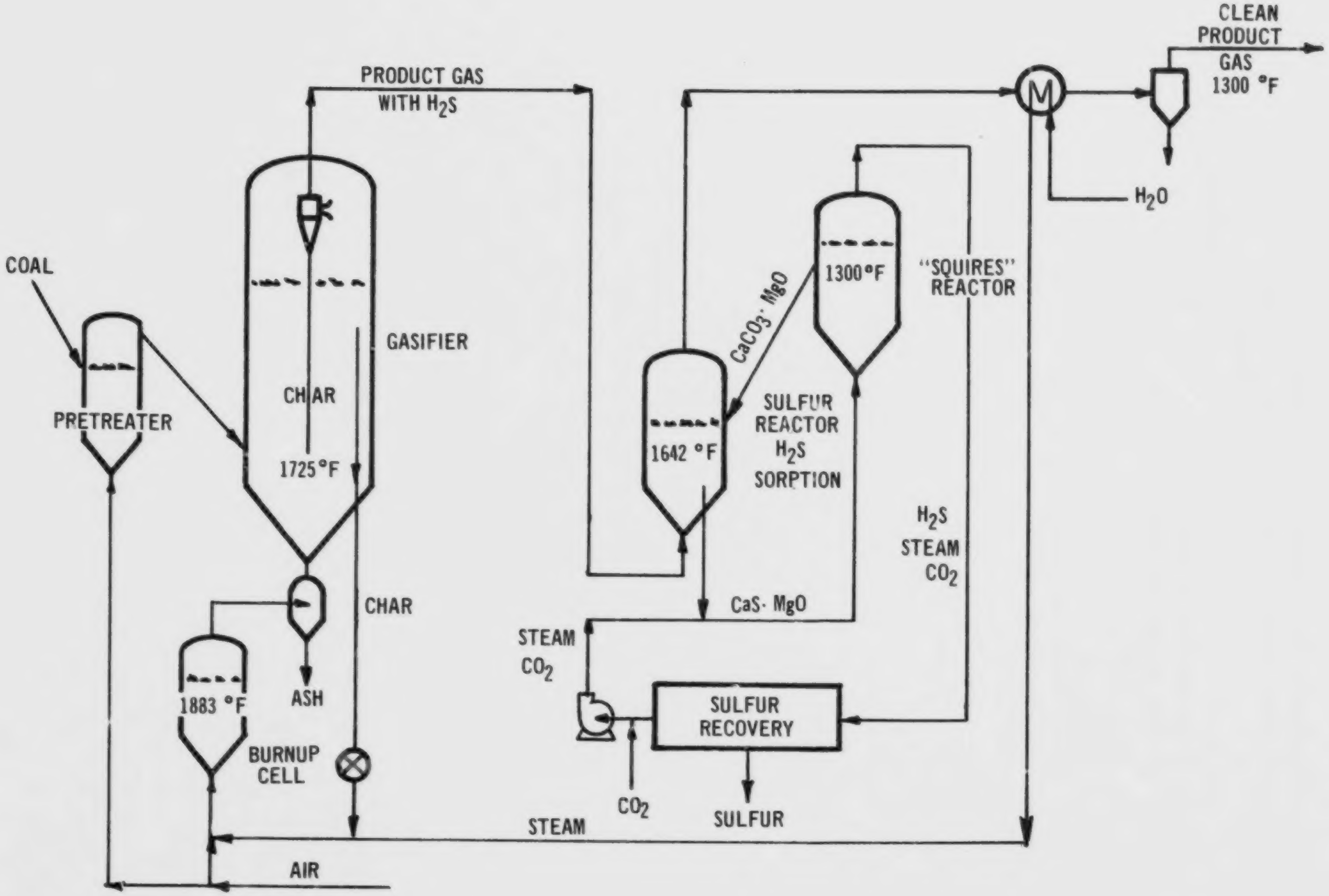

Figure 1. Two-stage fluidized-bed partial combustion process.

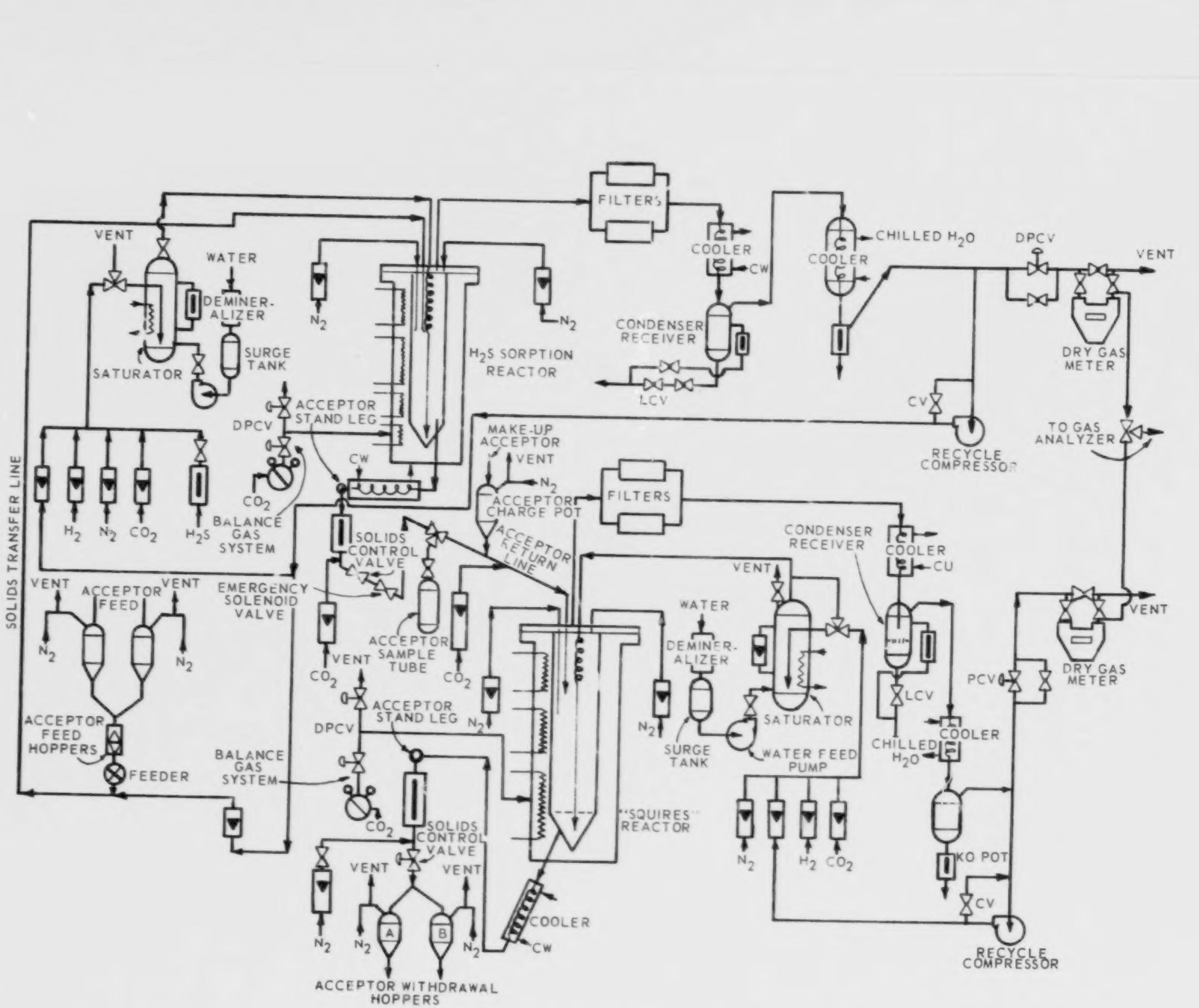

Figure 2. Flow diagram of new experimental unit.

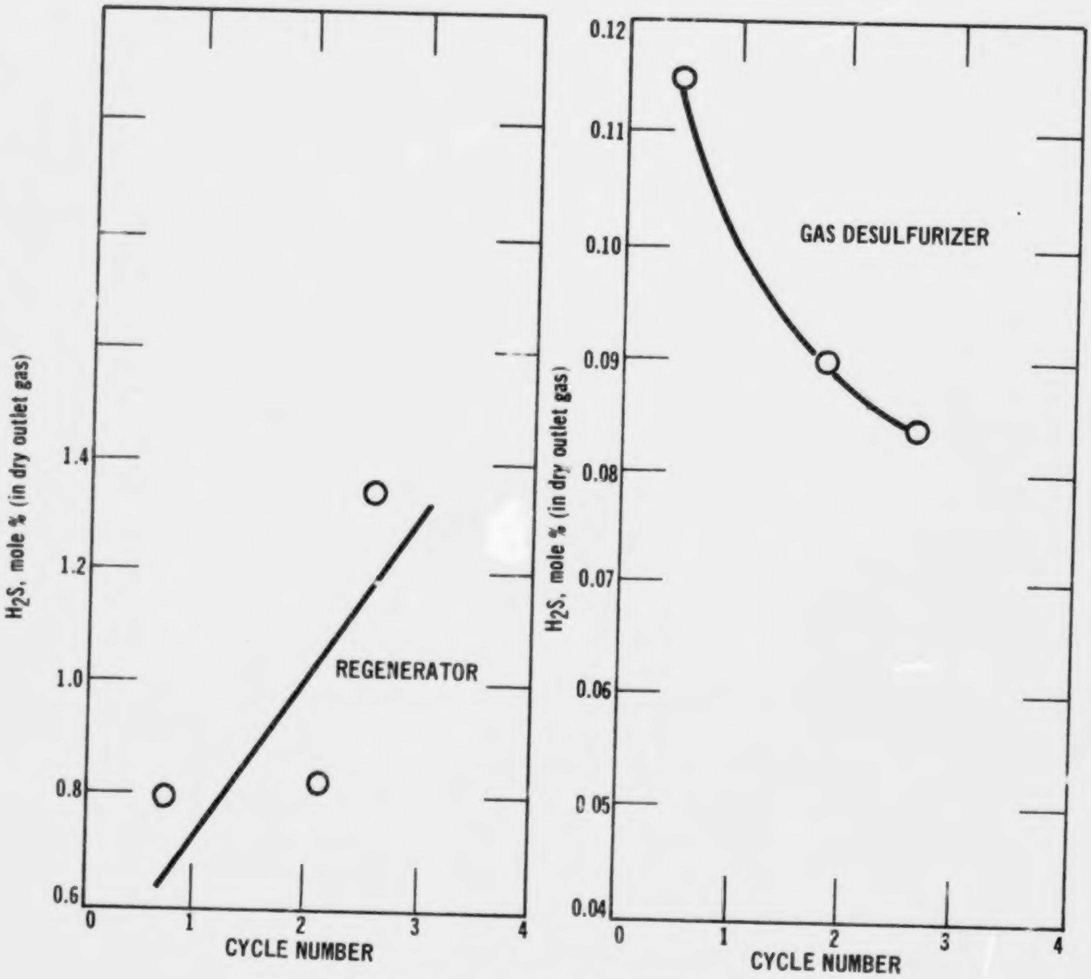

Figure 3. Run A4–tymochtee dolomite.

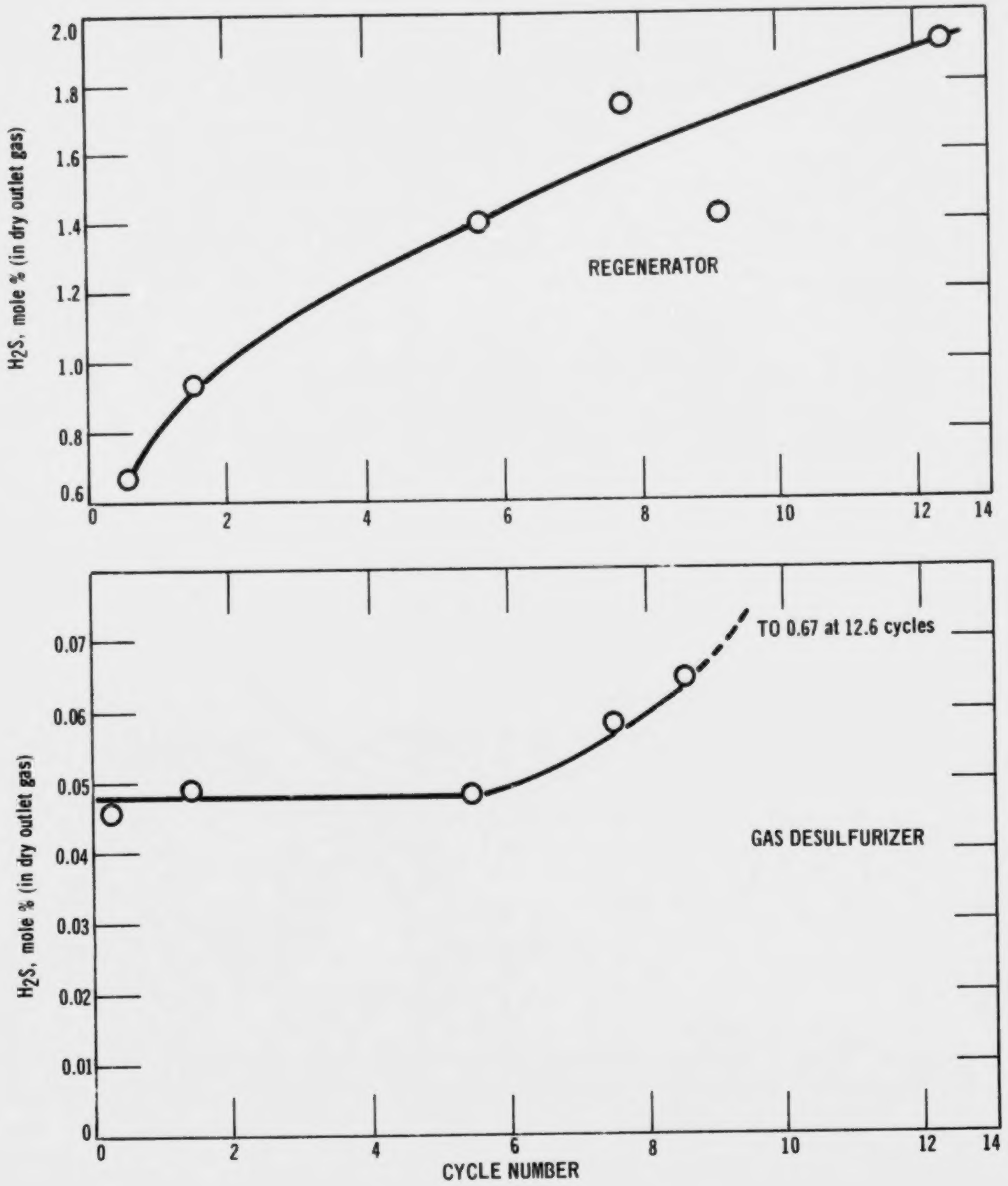

Figure 4. Run A7–canaan dolomite.

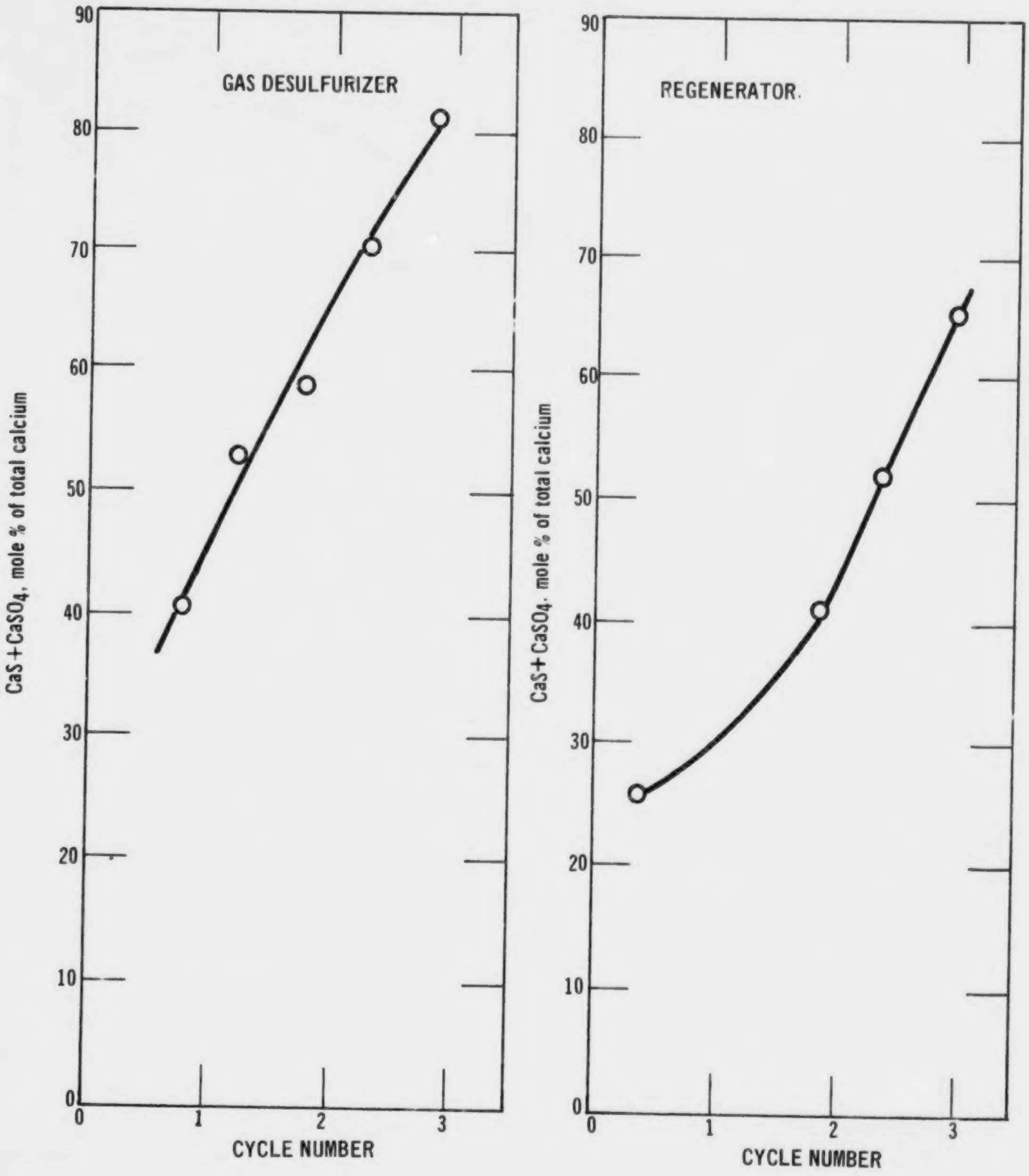

Figure 5. Run A4–molar sulfur content of exit solids.

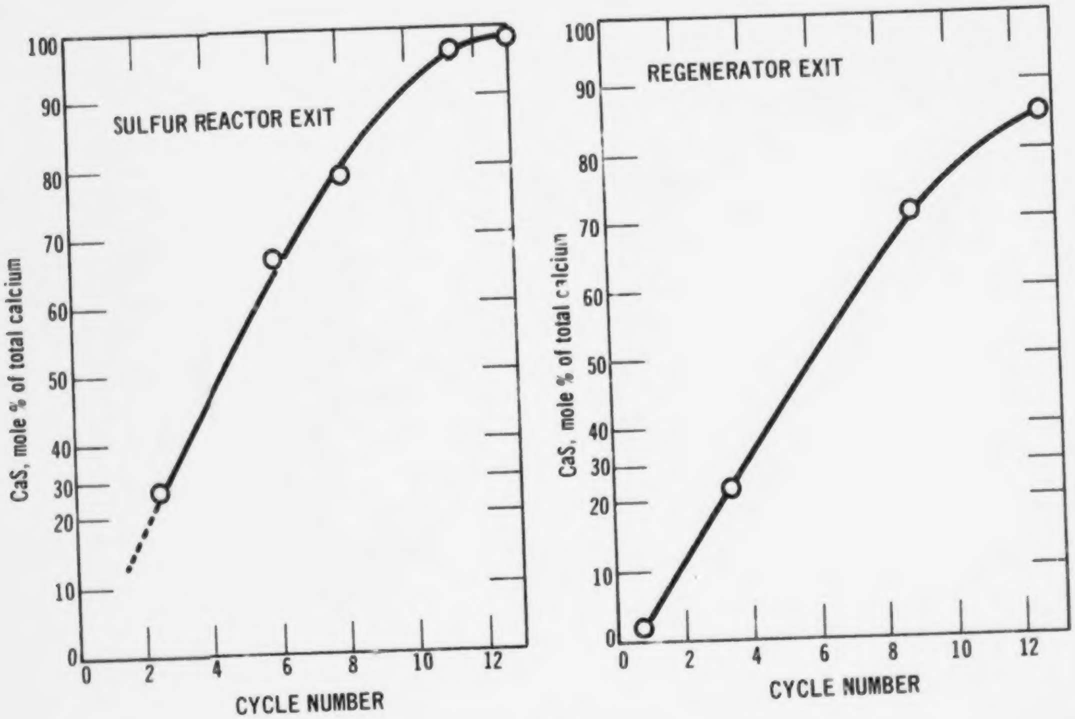

Figure 6. Run A7–CaS content of acceptor.

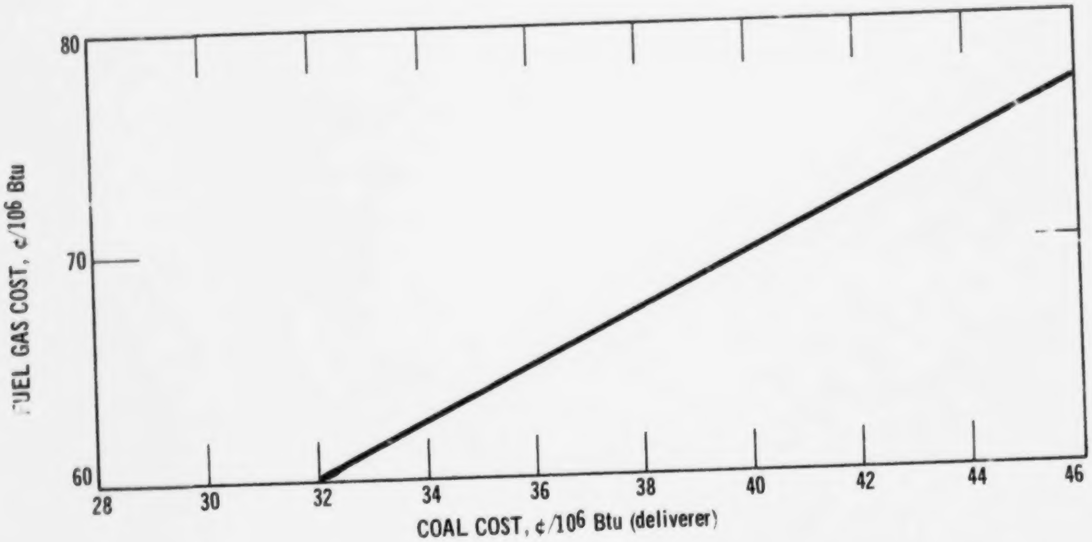

Figure 7. Fuel gas cost–effect of coal cost. Cost is based on fuel or fuel gas plus sensible heat content, 1976 operation.

SESSION IV:

Conceptual Designs and Economics

SESSION CHAIRMAN:

Dr. D.H. Archer, Westinghouse

1. SMALL-SCALE APPLICATIONS OF FLUIDIZED-BED COMBUSTION AND HEAT TRANSFER

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Research on fluidized-bed combustion has primarily been aimed at improved economics and anti-pollution measures for coal- and oil-fired power stations or for relatively large packaged boilers. At the last Hueston Woods Conference, however, the Keynote Address hinted that advantages might be gained from applying fluidized-bed combustion and heat transfer techniques even on very small systems.

In the two years since then, considerable progress has been made in research to identify and solve problems associated with small-scale application; a small development company has been started to exploit areas of likely commercial interest.

As some of the work involved may give feed-back to large boiler technology, this paper reviews the state of the art and gives data on combustion and heat transfer.

GAS-FIRED FLUIDIZED-BED COMBUSTION

Up to now, research at the University of Aston Mechanical Engineering Department has mainly concentrated on gas firing. That gas can be burned successfully in deep fluidized beds is well-known. Reference 1 describes Russian work. Reference 2 cites French research. The Coal Research Establishment (NCB) used gas combustion in the early days of fluid-bed coal burning work to investigate aspects of volatile burning. Most of this work was done in beds 1-ft deep or more; it required the use of relatively high pressure blowers, therefore necessitating either high-speed motors or some form of multi-stage rotors. In a practical plant the former solution would result in noisy appliances, while the latter incurs high manufacturing costs. A further disadvantage with deep fluidized beds when applied to small heat inputs is that the total surface area from which heat can be lost is high in relation to the throughput, so that beds with an L/D ratio of more than about 0.5 have to be surrounded with a high degree of lagging to prevent undue heat losses during startup.

Research was, therefore, initiated to find the minimum bed depth which would give stable and efficient combustion. Because of the very poor lateral gas mixing in fluidized beds, the idea of introducing separate gas jets into an air fluidized bed was discarded; the bed depth needed for complete mixing and good combustion would be too great unless an extremely large (and costly) number of gas jets were used. The experiments were conducted with pre-mixed gas/air mixtures fed through a porous ceramic distributor into a bed of silica sand (Figure 1). Provided that the gas/air mixture is initially within the flammability limits (2.2 to 9.5 percent by volume for the propane normally used) and the initial fluidizing velocity is less than the flame propagation speed, then such a bed can be ignited by simply lighting the gas/air mixture above the surface of the bed. Whether or not the system heats up to obtain controlled fluidized-bed combustion, however, depends upon having suitably sized solids particles, choosing an appropriate heat input rate for the system, and preventing undue heat loss at startup.

Various stages of startup on a bed with correctly chosen conditions (1-in. deep, 6-in. diameter, 18,000 Btu/hr input, 10 percent excess air) are as follows:

1. Air is blown through the bed at a rate which is around the incipient fluidizing velocity. The bed surface is hardly disturbed. A near stoichiometric ratio of gas is then admitted.
2. On ignition, the gas/air mixture burns at the top of the bed with a blueish flame, which dances around in an irregular pattern as it is not attached to any particular stabilizing system.
3. Particles which had been thrown up from the top surface of the bed into the flame and heated, now return to the bed and carry the heat down.
4. The gas/air mixture now becomes preheated as it passes into the bed and burns more readily at the surface with a distinct popping, roaring noise. The preheating causes the fluidizing velocity automatically to increase; it also causes more particles to be thrown up, which in turn maintains the rate of temperature rise in the bed. The flame structure modifies, and red-hot particles begin to tinge the colour of the flame.
5. When the bed temperature reaches 600°C, combustion begins which glows dull-red behind the reddish-blue flame. Noise level increases. By now the fluidizing velocity is some three times greater than the initial cold fluidizing velocity and more bubbling takes place. Combustion in the bubbles becomes more violent; mild explosions/detonations occur which tend to throw up far more particles from the surface of the bed.
6. When the bed temperature has reached 700°C, much of the combustion occurs in the bed; by 800°C only a small proportion of the gas burns at the top. The noise level is now at a peak, probably indicating that the combustion is occurring after the gas/air mixture has formed itself into bubbles within the fluidized bed. The surface splashing of particles has decreased, which indicates that although it is suspected that combustion occurs in the bubbles, these bubbles are further below the surface when combustion occurs than in the case of the 600°C bed.
7. By 900°C the noise level is reducing; at 1000°C the level has dropped several decibels below its maximum level. The ultimate temperature level reached depends mainly on the gas and air input to the bed. Temperatures up to 1200°C can be readily maintained with silica sand, but particles with higher fusion temperatures are needed to go much beyond this temperature because of sintering. The noise level is reduced in deeper beds over 800°C and disappear above 880°C.

Figure 2 shows a typical heating up rate for a shallow bed combustor; Figure 3 shows the noise level spectrum emitted from the bed at various temperatures. It is believed that lower noise levels occur at the higher temperatures because combustion is extremely rapid in the first few millimeters of the bed before the gases have time to form into bubbles. Thus, the likelihood of detonations occurring is reduced because the combustion is quenched by the large number of particles present.

STABILITY

Once a temperature above 800°C has been reached, the system can be operated over a wide range of gas/air mixture strengths well outside the normally accepted limits set by flame propagation phenomena. The fluidized bed operates as a very effective pre-heater, bringing the incoming gas up to bed temperature within the first few millimeters of the bed.

Provided the external heat losses from the bed are small, achieved by lagging and by placing reflecting surfaces above the bed to radiate the heat back down to the bed, extremely weak gas/air mixtures can be burned.

Combustion can be maintained with extremely shallow beds (below 0.5-in.), but it is not yet clear whether or not complete temperature equilibrium between the exit gases and the solids is achieved. With very shallow beds, some combustion probably takes place after the gases leave the bed. For beds 0.5-in. deep and above the exit gases appear to be more or less in temperature equilibrium with the bed, and combustion efficiency is excellent with CO/CO_2 ratios dropping below 0.002 (a factor of ten better than the standards insisted upon by the UK Gas Council for domestic appliances).

Because of the quenched low temperature combustion it was expected that NO_x production would be low. This has been borne out by samples drawn through Dager tubes measuring $NO + NO_2$ which indicated less than 5 ppm in the exhaust gases. These figures represent a considerable reduction compared with emission from normal flames.

It is interesting to note that the combustion intensity of these beds is in the region of one million Btu/ft^3-hr based on the full depth of the bed. As it is likely that most of the combustion takes place in the bottom half of the bed, the actual combustion intensity must be at least twice this rate. A similar bed, but with heat transfer by direct contact between the particles and cooling surfaces, can be operated at more than twice the above heat release rate; figures of 3×10^6 Btu/ft^3 -hr have already been achieved.

The ease of operation of this form of gas "burner" in the temperature range of 800 to 1200°C, combined with the excellent heat transfer which occurs when small objects are placed in the bed makes possible the use of such furnaces for laboratory, workshop, and factory metallurgical processes, e.g., hardening, annealing, heating small billets prior to forging etc. Figure 4 shows a typical heating curve for a 3-in. long x 1/4-in. diameter alloy steel bolt immersed in a fluid-bed combustor operating at 960°C (the extra heat absorbed during the transformation zone is clearly seen). With these possibilities in mind a small company, Fluidfire Development Limited, has designed and built a range of furnaces:

1. 6-in. diameter x 1-in. deep self-contained units for laboratory investigations and demonstration work.
2. 6-in. diameter x 6-in. deep units again for laboratory work which contain a two-stage blower or can be used from a shop air supply.
3. An 8-in. diameter x 8-in. deep metallurgical furnace with automatic temperature regulation for use in small quantity hardening and annealing work.
4. A larger 12-in. diameter x 12-in. deep metallurgical unit, again fully temperature controlled, shown in Figure 8.

The metallurgical furnaces which can operate over a temperature range of from about 700 to 1200°C can replace traditional salt bath furnaces for hardening and tempering a wide range of materials. The fluidized furnace has the following advantages:

1. Higher operating efficiency allows lower fuel costs per pound of metal processed.
2. The cost of special salts and the difficulty of handling and disposing of the spent salt are eliminated.
3. The furnaces have a short startup time and therefore can be switched off overnight.
4. The atmosphere in the heating zone can be adjusted to suit the requirements of treatment. It is usually made to be reducing.
5. The furnaces can operate over a wide range of temperatures and with slight modifications can be changed to carburising duties in which case they eliminate the use of cyanide with all its attendant safety and disposal problems.

6. All types of steels may be heat treated.

The performance of these units with respect to operating cost, heating rates, and oxidising rates at various temperatures and the hardness achieved under various conditions is described in reference 3.

A special unit has also been designed to operate in line with automatic continuous production of hardened steel components. This unit uses a continuous wire mesh belt to support and convey articles through two separately controlled fluidized- bed combustors: the first higher temperature combustor is the heating zone; the second combustor, running at a precisely controlled temperature, allows a short soaking period and ensures that the articles are fed into the quenching system at the correct temperature.

RADIANT HEATERS

Shallow-bed combustion systems of this type are interesting in their own right since they are a new way of making more effective radiant gas heaters. Normally, radiant gas heaters rely on convective heating of ceramic plaques by very hot gases and the re-radiation of the heat from the plaques to the surroundings. The exit gases are at a substantially higher temperature than the radiation surfaces; the overall effectiveness is generally such that only 25 to 35 percent of the input heat is radiated.

The immense surface area exposed to the combustion gases in a fluidized bed allows the temperature differential between the gases and the solids to be negligible. Thus, a bed operating stoichiometrically at 1000°C will radiate approximately 50 percent of its heat away from the bed. The operating temperature/radiation efficiency of an ideal bed (i.e., perfect burning of the air and gas before leaving the bed and thermal equilibrium between the gases and the solids) can be readily calculated. The radiation efficiency is given by the expression

$$n = \text{radiation} = 1 - \frac{h_t}{H}$$

where: h_t is the enthalpy of the exhaust gases at the bed operating temperature, and H is the calorific value of the fuel burned.

There are only two sources of heat loss from the bed, the enthalpy of the exhaust gases and the radiation from the bed. The external convective heat losses are negligible if the height of the containment wall is low. Any heat directed downwards to the distribution plate is returned to the combustion bed as preheat in the gases.

Figure 5 plots the radiation efficiency versus bed temperature for various gases. It will be noted that the radiation efficiency for stoichiometric mixtures is not very different for the various gases and is not a function of the bed emissivity which only affects the rating of the bed per unit area. Figure 6 shows how the heat input and the radiant output of a 6-in. diameter shallow-bed combustor varies as the temperature of the bed changes for stoichiometric gas/air mixtures. The full curves correspond to a bed of unit emissivity and the dotted curves to a bed of emissivity 0.7.

It will be noted that as the bed operating temperature is lowered by reducing the gas/air volume fed to the bed, the radiation efficiency increases. Above about 800°C the small fluidized beds which have been produced perform very nearly as shown provided that due account is taken of the effective emissivity from the bed surface. It was originally expected that the emissivity of the granulated surface of a gently bubbling fluidized bed would act very nearly as a black body for most solids. But this does not appear to be the case; the emissivity for silica sand is approxmately equal to that of silica sand itself.

Below 800°C the radiation effectiveness is not as good as predicted; either thermal equilibrium is not established or some gas is leaving the surface unreacted.

Although the emissivity of a gently bubbling bed falls into line with the individual particle emissivity, the overall effective radiation from a fluidized-bed combustor using very fine particles may be significantly higher than this. As might be expected, the influence of a particle cloud above a fluidized bed materially affects its radiation characteristics. This effect exists because the particles during their stay in the gas space above the bed radiate their heat very rapidly, cool down to a temperature below that of the off-gases, and thereafter tend to act as a second-stage cooling medium for the gases leaving the bed. Thus, it should be possible to operate a fluidized-bed combustor at a temperature which is significantly more than the temperature of the gases leaving the system. In this case, the cloud of particles above the bed shows a duller colour than one would normally expect from a bed operating at the same temperature. Research into this phenomena is underway at Aston.

It will be seen from Figure 6 that the heat output of a 6-in. diameter bed operating at 1000°C is approximately the same as that of a traditional British radiant gas fire. This has led to the concept of using shallow fluidized-bed combustors as room heating devices. The constantly varying pattern of the fluidized bed, coupled with the similarity to the open coal fire, was thought to offer an attractive alternative to the traditional fire. Thus, self-contained units incorporating brushless electric motors, fans, controls, and safety devices are now being developed by Fluidfire Development Limited in order to assess the potential of such appliances. Although much more work is still needed to satisfy stringent safety requirements, many problems inherent in such a radically new system have been overcome, and there appears a fair chance of success.

NOISE

One of the requirements for room heaters is that they should not generate a great deal of noise, and the UK Gas Council recently proposed a standard of acceptable noise levels shown by the dotted line on Figure 3. It will be seen that the noise level from an open radiant bed is slightly higher than the permitted noise levels. Thus, to quiet the fire somewhat and also to lessen the danger of having a completely open fire, a glass screen could be incorporated in front of the fire. Tests with ceramic glass show that this reduces the noise level to well below the acceptance levels and ensures that clothing cannot be ignited by direct contact with the fire.

BOILER APPLICATIONS

It is obvious that if a radiant fluidized-bed combustor operating at a temperature level of 800 to 900°C is surrounded by a water jacket, then 55-60 percent of the heat input will be radiated to the water walls, even if the fluid-bed particles do not come into contact with the walls. However, the rating of such a boiler would be relatively low—a 6-in. diameter bed having an output of something like 15,000 Btu/hr. Hence, the unit would not be particularly compact and could not be considered as a viable commercial alternative to the highly rated gas-fired boilers which are now being produced.

If an attempt were made to place heat transfer tubes in the fluidized-bed combustor in a similar manner to the way in which large fluidized boilers have been designed, then the simple startup procedure described earlier would not be effective, and compartmentation of the bed for startup purposes would be necessary. For small-scale appliances, this would be prohibitively costly. An alternative was sought; the idea of locating heat transfer surfaces just above and around the settled fluidized bed was formulated and has been successfully developed. This solution relies on the principle that the expansion of a shallow fluidized bed (as a percentage) is very high compared with a deep fluidized bed. Thus on startup with cold air the bed expansion is very small, and the particles do not contact the heat transfer surfaces. If a gas/air mixture is then lit above the bed, the bed heats up in a similar fashion to the radiant bed described above; but by the time a temperature of 700 to 800°C has been reached, the fluidizing velocity is some three to four times the initial velocity and the bed has expanded to contact the heat transfer surface. Further contact takes place by virtue of splashing of particles from the bed upwards and sideways.

For beds of small output (up to 100,000 Btu/hr it is sufficient to surround the bed with a water-cooled wall which is insulated from the settled bed.

The direct contact between the particles and the cooling surfaces allow the heat input to be two to three times that which could be sustained in a bed which was only cooled by radiation. The principle could still hold good for larger outputs, but in this case some additional heat transfer surface would have to be placed in such a position that it contacted the expanded bed and received splashing heat transfer.

Because the fluid-bed combustor will not operate very satisfactorily below 800 °C, and even if we could exploit futher the principle of particle cloud radiation to cool the off-gases to below bed temperature, the overall efficiency of the combustion system in transferring heat to water would be too low for domestic central heating systems. Some form of second-stage heat recovery is therefore necessary. The incorporation of convective heat transfer surfaces would leave the system with many of the disadvantages of normal systems, e.g., large heat transfer volumes or the use of high extended surfaces with the possibility of condensation and corrosion troubles. A second-stage shallow fluidized bed was therefore incorporated above the combustion bed of a trial 40,000 Btu/hr laboratory unit which was supplied by air from an external source. With a 1-in. bed depth and an 8-in. diameter, the heat transfer area of about 1/6 square foot around the periphery of the bed allowed the existing gases to reduce in temperature to 400°C, giving an overall efficiency of about 80 percent.

The addition of a number of thick fins to the walls increased the surface area to 1/2 square foot and enabled the gas temperature to be reduced to less than 250°C, giving an overall efficiency of about 87.5 percent.

An important point is that these high efficiencies are achieved without operating with metal temperatures below about 110°C. This is possible because of the very high heat transfer coefficients between the fluidized solids and the metal surfaces. These coefficients result in the significant advantage that condensation of the exhaust gases on the fins does not occur and corrosion is eliminated. This is not the case if highly extended surfaces are used in normal convective heat transfer.

Figure 9 shows a prototype domestic fluid-bed boiler incorporating blowers, controls, safety cut-outs, etc. This latter unit is now carrying out endurance trials to determine the rate of loss of particle mass during extended running.

With regard to particle life, experiments have already been conducted at room temperature using sand in a fluidized bed operated at 2 ft/sec; no measurable attrition loss occurred in 500 hours. Whether or not the continual heating and cooling of particles in a combustor with the consequent thermal shocking will produce more severe attrition is as yet unknown.

Apart from the very effective heat transfer in the second-stage bed, a further significant advantage is that it produces an extremely effective silencer for the combustion system. Provided, therefore, that particles to resist degradation can be found, it appears likely that this type of approach can provide central heating units with the following characteristics:

1. Very low NO_x emission (<5 ppm).
2. Very low CO/CO_2 ratios.
3. Lower aldehyde formation than for normal flames due to the low temperature combustion.
4. Efficiencies as high as 90 percent without significant extra cost and with no fear of condensation or corrosion.
5. Very compact plant with overall ratings of 100,000 Btu/ft^3 obtained (including fans, motors and gas controls).

6. Use of well-established cast iron techniques which are cheap and produce long life, low maintenance units, to produce boilers.
7. Easily adjustable units for all gases and Wobble irrelevant number and flame speeds.
8. Good turn down ratio of units without losing efficiency.
9. Units readily developed to burn oil and with a little development could possibly burn solid fuel, (but could still be switched back to gas easily).
10. Application of techniques to visible fire/back boiler systems providing a central focus in the living area as well as full house heating.

SHALLOW BED HEAT TRANSFER

Early studies by the Central Electricity Generating Board showed that uneconomical high pressure drops would be incurred if an attempt were made to use plain tubes in a fluidized bed as a straightforward heat recovery system (i.e., one not using a combustion reaction). Early work by Petri et al.[4] showed that if a tube were provided with an extended surface finning system with an area 15 times that of the base tube, then the overall heat transfer would be increased approximately six-fold, i.e., an effectiveness of 40 percent. As the overall tube diameter would not be more than doubled by employing the fins, the net effect would be a far greater heat flux per unit volume of bed. Thus, the pressure drop penalties when using extended surfaces would be significantly reduced.

Following up this idea, a new form of extended surface heat exchanger was built, and some preliminary results were presented to the Second International Conference on Fluid-Bed Combustion. These preliminary results were obtained with the extended surface systems in a comparatively deep bed with fairly large particles. Further work showed that if this particular arrangement of vertical fins was placed low down in a fluidized bed, its performance was unexpectedly higher. Figure 7 shows the performance of vertical-finned extended surface tubing operated in a very shallow fluidized bed compared with that for 1-in. tubing and with the results of Petri et al. The graph plots the bed-to-metal heat transfer coefficients against particle size. It will be noted that the shallow bed results lie above the generally accepted heat transfer coefficient line. These results do not necessarily imply that the vertical surfaces have an effectiveness of over 100 percent, although they may in some instances, but do show that shallow-bed performance is superior to that of deep beds, to which most of the world's data on heat transfer relate.

The reason for the superiority of the particular configuration of vertical surfaces in shallow fluidized beds is believed to be the absence of large bubbles in the system. The absence is partly because the vertical surfaces prevent lateral mixing of the gases which again restricts bubble formation. It is interesting to observe that when these extended surface tube bundles are placed in a shallow bed they do not appear to disturb the bubbling pattern.

A further possible explanation for the improved heat transfer is that the viscosity of shallow fluidized beds varies almost directly as the bed depth. Shear stress/shear strain data derived from a Stormer-type viscometer with a hollow cylindrical rotor is given in reference 5. Because of the better fluidization in shallow beds, the resistance to shear of the fluidized solids is much less. As we know that fluidized-bed heat transfer depends upon the rate of exchange of particles at the heat transfer surfaces, it would be logical to expect that heat transfer would improve if particle mobility improves. Thus, shallow beds would be expected to be superior to deep beds from a heat transfer aspect.

Coupling extended surfaces with extremely shallow beds has been patented with the concept that we need no longer regard fluidized beds as isothermal devices. This enables the overall thermal effectiveness of a fluidized bed to be improved by the correct design of the heat transfer bundle/distributor unit.

As pointed out at the last conference here, in contrast to conventional convective extended-surface heat transfer systems where the improvement in heat fluxes per unit volume is accompanied by a higher pressure drop, the use of fluid-bed extended surfaces increases the heat flux per unit volume and decreases the pressure drop. The pressure drops in some of the shallow-bed units we have investigated are so low that the system can now compete favourably with normal convective heat transfer, even for gas turbine waste heat recovery where low pressure drops are of paramount importance. It is contemplated that a two or three stage fluid bed can be operated with an overall pressure drop of less than 12 inches water gauge.

Theoretical and experimental studies of the mechanism of heat transfer between the fluidized solids and the vertical fins and along the fins themselves are underway; a vast amount of data has been obtained for various proprietory extended surface tubing as well as for specially designed fin/tube arrangements. These data, which are at the moment being written up for presentation in the near future, are sufficiently complete to allow an economic appraisal of various forms of heat exchangers to be made. Further advances in performance are expected when a better appreciation of the various phenomena is acquired.

It is suggested that for fluidized-bed boilers or for combined gas turbine steam cycles where steam is generated in the high temperature exhaust from the gas turbine, there is already a case for investigating the use of shallow fluidized-bed extended-surface systems instead of normal convective heat transfer.

Preliminary trials of a single stage unit picking up waste heat from a diesel engine have been very encouraging. Heat transfer coefficients of the bed which was completely covered by vertical fins were just as good as for the original laboratory unit. The fluidized solids and the fins became coated with carbon, suggesting that there may be some possibility of enhancing this effect to reduce pollution from the sub-micron carbon in the exhaust gases. The unit ulso acted as a very effective silencer.

FUTURE RESEARCH AND DEVELOPMENT

Initial experiments into burning distilate oil in fluidized beds have been successful; it seems likely that a dual fuel gas/oil system can be developed quite quickly. The unit would have no higher pressure drop than the existing gas-fired unit and, therefore, would have advantages over normal pressure jet burner furnaces with regard to fan pressure and noise levels. Its pollution control level would be far superior. It is expected that the unburnt hydrocarbons will be much reduced compared to normal oil flames.

Research on small-scale solid fuel-fired units has also started; there appears to be no insuperable problems in producing a shallow open-hearth solid fuel fire giving radiation outputs of over 60 percent. A high-efficiency domestic fluid-bed solid fuel-fired boiler also appears to be a practical proposition

The extension of these ideas into the field of packaged boilers is underway; it is expected that economically viable units can be developed.

In addition to the low pressure, hot water boiler developments, studies of high temperature, high pressure steam systems indicate that such units can be designed to startup and operate satisfactorily and economically. These units would employ high temperature alloy tubing capable of operating dry during startup, thus avoiding heat losses to the cooling system during startup. A unit capable of producing steam for a 150-hp engine would have a diameter of approximately 2 feet with a combustion bed 6-in. deep followed by a further 6-in. deep economiser. With shallow fluidized-bed extended surface systems the bed depths would be even smaller.

CONCLUSIONS

Domestic fluidized-bed combustion/heat transfer systems which are no more expensive, are just as compact, and have far less pollution than their conventional counterparts have been developed.

Endurance and reliability trials are in progress so that by the end of 1972 we should be able to assess the full potential of gas-fired fluidized-bed combustion as applied to small-scale boilers. The results so far suggest that additional work should be undertaken on oil- and coal-fired systems.

Fluidized-bed combustion and heat transfer lends itself well to metallurgical heat treatment processes in which the antipollution aspects combined with rapid processing have been shown to lead to environmental and economic benefits. Batch processing furnaces are now available, and in-line continuous furnaces show distinct promise.

The development of shallow bed, extended surface heat transfer systems promises to open up a completely new field in heat recovery and could help to reduce costs and space requirements in many types of plant.

It appears, therefore, that fluidized-bed combustion and heat transfer techniques can be usefully employed for antipollution measures over the whole range for units having an output of a bunsen burner up to extremely large power station sizes; in many cases it will be accompanied by economic benefits.

REFERENCES

1. Ukilov, V. M., G. K. Rubtsov, and A. P. Baskakov. Gas Combustion in a Packing under a Fluidized Bed. Gazovaja Promyshlennost. 5, 1969.
2. Tamalet, A. J. Application of Fluid Bed Heat Transfer to Metallurgical Processes. Inst. Chemical Engineering Symposium Series. In: Proceedings Symposium on Chemical Engineering in Iron and Steel Industry, pp. 105-114, 1968.
3. Virr, M. J. and R. Reynoldson. Heat Treatment in Fluidized Beds. Industrial Process Heating.
4. Petrie, J. C., W. A. Freeby, and J. H. Buckham. Bed Heat Exchangers. Chemical Engineering Progress, 64(7), 1968.
5. Botterrill, J. S. M., M. Van der Kolk, D. E. Elliott, and S. McGuigan. The Flow of Fluidized Solids. Powder Technology, 6:343-351, 1972.

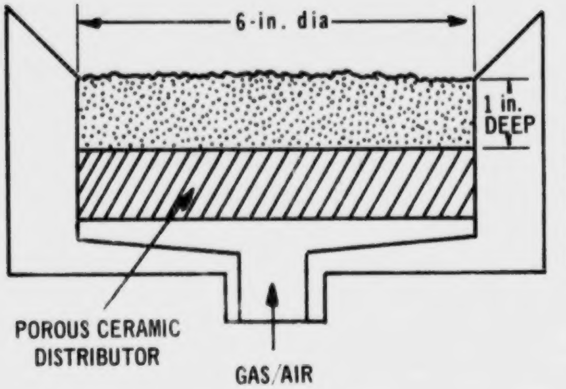

Figure 1. Radiant cooled shallow fluidized bed.

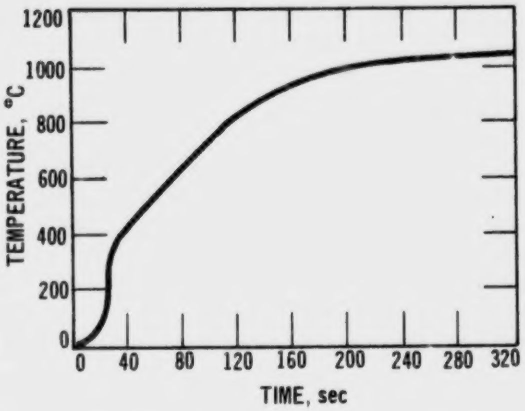

Figure 2. Start-up temperature/time sequence.

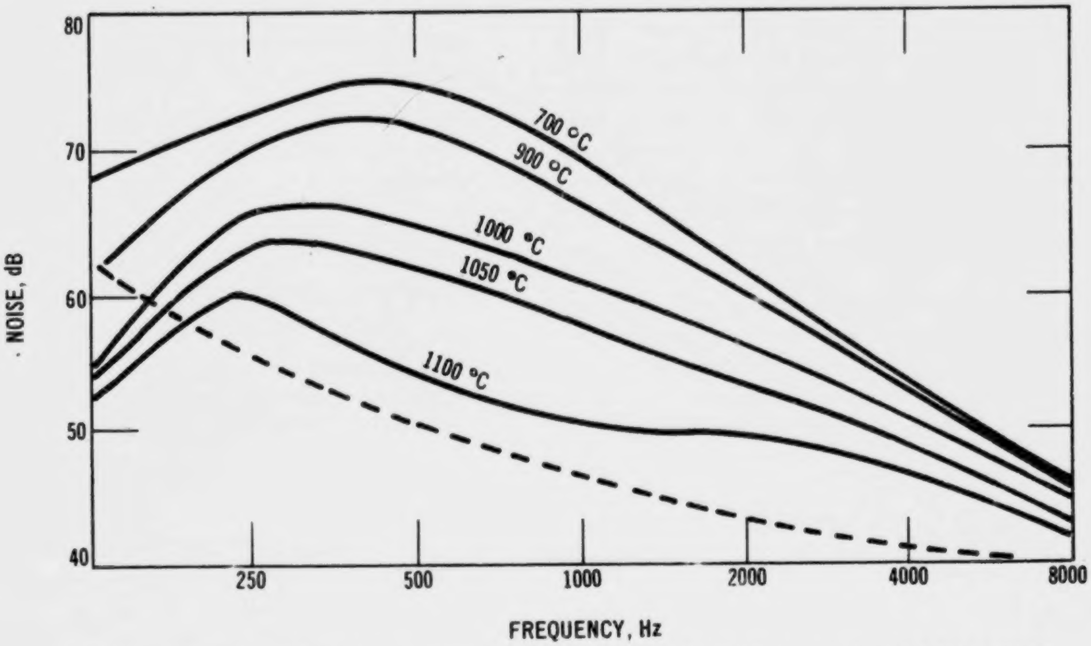

Figure 3. Noise levels 2 ft away from radiant bed (including unsilenced fan noise).

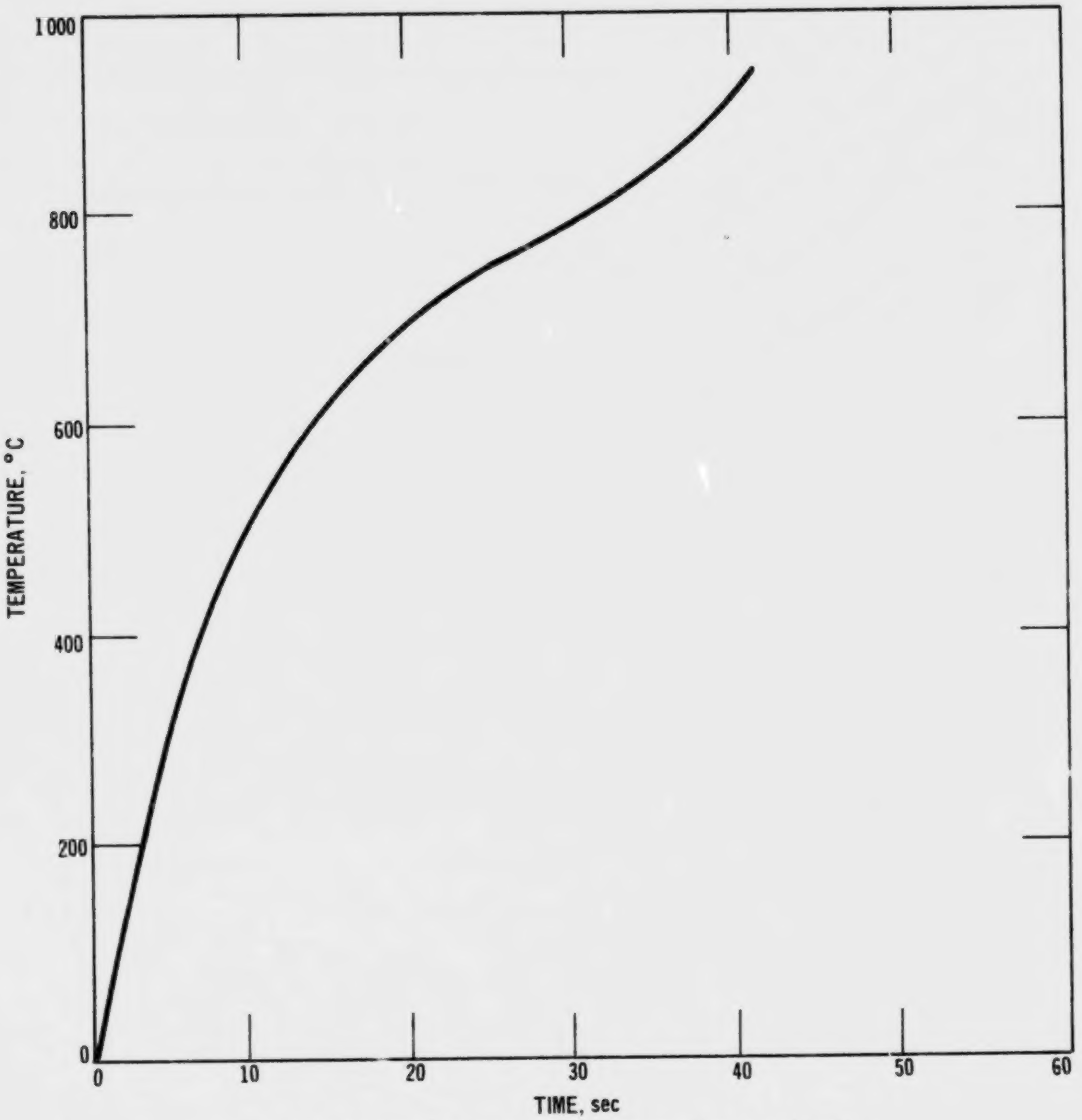

Figure 4. Heating of ¼-in.-diameter alloy steel bolt.

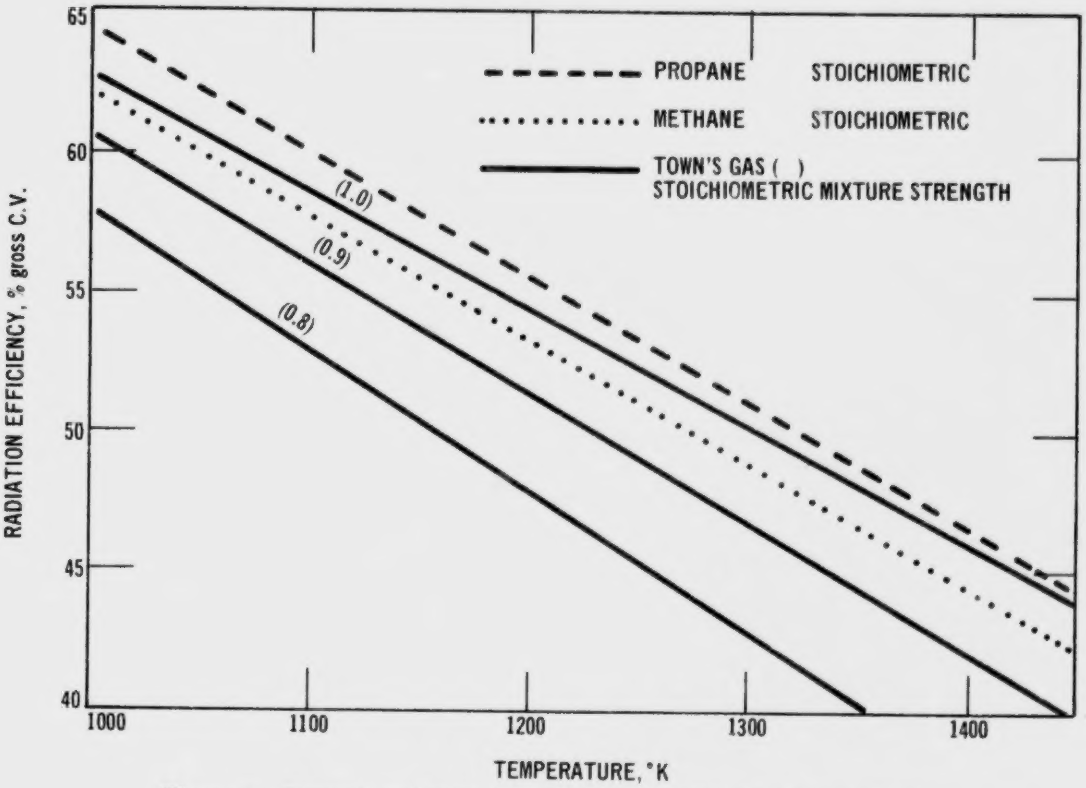

Figure 5. Percent radiation from shallow fluidized bed combustors.

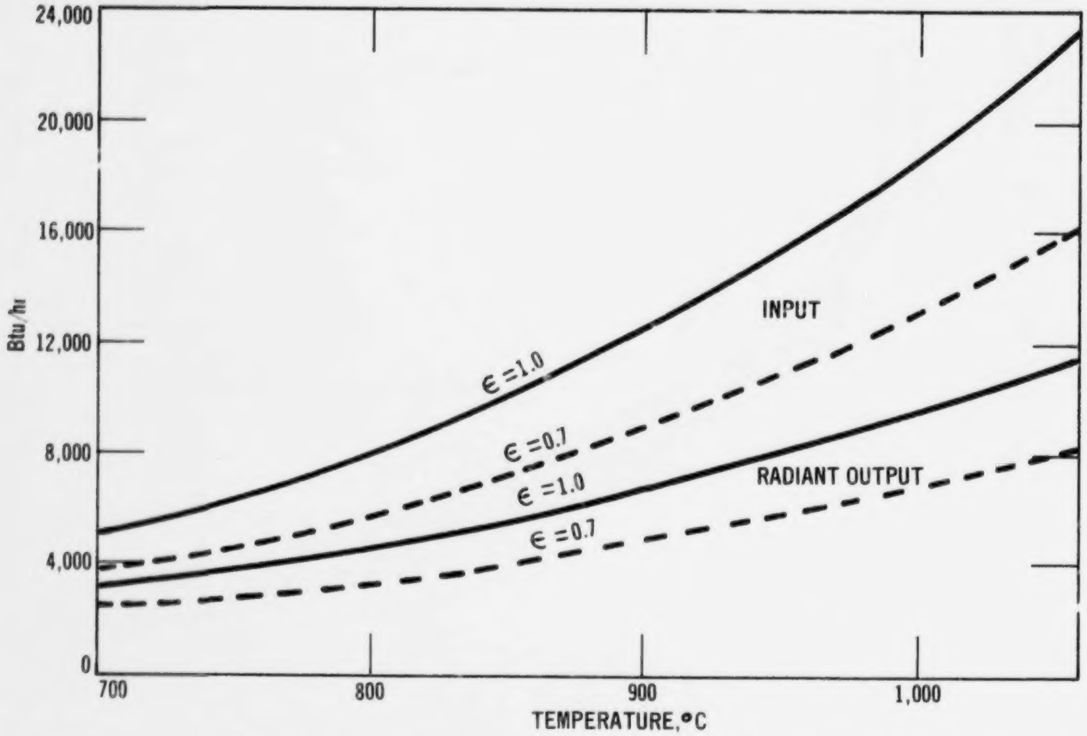

Figure 6. Input and radiant output from a 6-in.- diameter shallow bed combustor (based on gross calorific value).

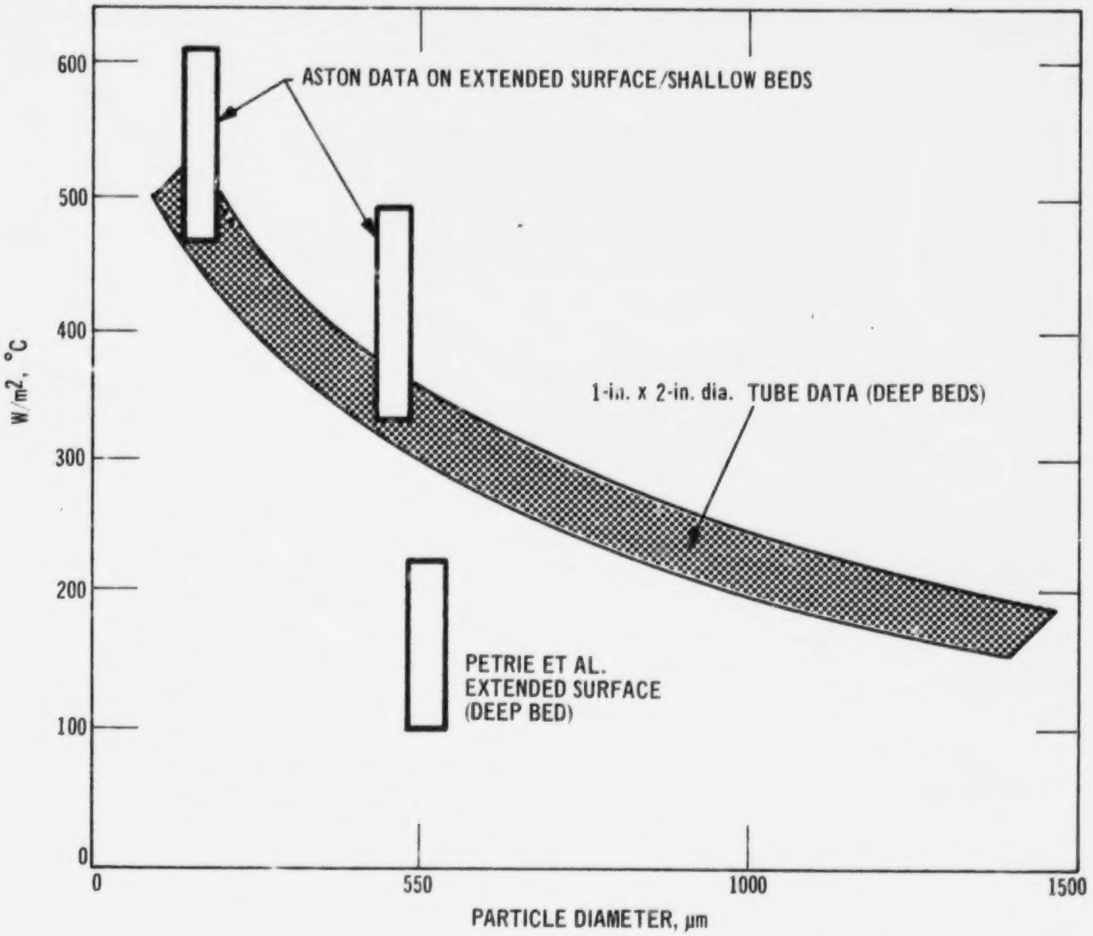

Figure 7. Comparison of shallow/deep bed heat transfer.

2. FLUIDIZED-BED COMBUSTION UTILITY POWER PLANTS—EFFECT OF OPERATING AND DESIGN PARAMETERS ON PERFORMANCE AND ECONOMICS

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ABSTRACT

Pressurized fluidized-bed boiler power plants have the potential to meet SO_2, NO, and particulate emission standards at energy costs 10 percent below conventional plants with wet scrubbing. This paper analyzes the sensitivity of the operating and design parameters selected for the plant design on plant performance and economics. Results show that the plant costs and performance are essentially invariant with projected changes in operating and design parameters—2.5 percent change in energy cost. The concept has the potential for achieving plant efficiencies of ~45 percent.

INTRODUCTION

A pressurized fluidized-bed boiler power plant has been designed using state-of-the-art power generation equipment.[1,2] Performance, costs, and pollution abatement were projected for the system. The results show the concept has the potential to meet SO_2, NO, and particulate emission standards and may reduce energy costs 10 percent below a conventional plant with stack gas scrubbing.

Operating conditions and design parameters for the pressurized boiler were selected based on an evaluation of available data, power cycles, and alternative boiler concepts. It is important to know how sensitive the operating and design parameters selected for the base design are to the plant economics. An understanding of the effect of changes in the proposed design on plant cost will provide a basis for evaluating current pressurized fluid-bed combustion pilot plant data, planning experimental programs, designing the development plant, and understanding the economic margin for solving technological problems.

The sensitivity analysis evaluates the effect of the following variables on plant design, cost, and performance:

- Fluidized bed boiler operating conditions
  - Bed temperature
  - Fluidizing velocity
  - Excess air
  - Pressure
- Fluidized bed boiler design
  - Heat transfer surface—configuration, heat transfer coefficient, and materials
  - Module capacity
- Particulate carry-over from the boiler
  - Loading
  - Size distribution
- Power plant equipment operating conditions
  - Gas turbine inlet temperature
  - Steam temperature and pressure

The evaluation is performed by considering each variable separately; it is general in order to permit the coupling of different effects to assess alternative designs. It is performed to indicate relative effects of variable changes in plant costs relative to each other and the total plant cost.

BASIS FOR SENSITIVITY ANALYSIS

The basis for the sensitivity analysis is the boiler and plant design developed by Westinghouse under contract to EPA.[1,2] The power plant cycle is shown schematically in Figure 1. The plant subsystems included in this sensitivity analysis are enclosed within the broken lines. The pressurized boiler was designed by Westinghouse and Foster Wheeler and is shown schematically in Figure 2. The preliminary boiler design was for a nominal 300-MW plant. The boiler design consists of four modules; the modularized design provides for a maximum of shop fabrication and turndown requirements. Each module includes four primary fluidized-bed combustors, each containing a separate boiler function—one bed for the pre-evaporator, two beds for the superheater, and one bed for the reheater. Evaporation takes place in the water walls. All of the boiler heat transfer surface is immersed in the beds, except for baffle tubes above the bed to minimize particle carry-over. Each module contains a separate fluidized bed or carbon burn-up cell to complete the combustion of carbon elutriated from the primary beds. The philosophy used to design the boiler was to maximize shop fabrication. Thus, the 300-MW plant utilizes boiler modules which can be completely shop fabricated. From roughly 300 to 600 MW, the boiler can be partially shop fabricated—the pressure shell being too large for rail transport. Larger plants, utilizing the four-module concept, would be field erected.

The operating conditions and design parameters for the boiler and the power cycle are summarized in Table 1. The power plant performance and economics were based on these specifications. The cost breakdown for the fluidized-bed steam generator is summarized in Table 2. The fluidized-bed boiler design was scaled to 600-MW capacity and the costs estimated. These costs are summarized in Figure 3. A breakdown of the power plant equipment costs for a 635-MW plant is presented in Table 3. Limestone or dolomite regeneration is not included in this analysis. Thus the costs presented are for a once-through system. The energy costs used for this analysis are also presented in Table 3. The costs of a conventional plant with wet scrubbing on the same basis are also indicated.

The following assumptions are made for the sensitivity analysis:

1. The plant concept maintains the four module with two modules per gas turbine concept.
2. Coal feed rate is maintained constant for each variable analysis. Thus the coal feeding and handling system is assumed to remain unchanged. This may not be completely true if the bed area is changed significantly and the number of feed points increased or decreased. The cost of the coal feed system is considered if the bed design is altered.
3. Structural and erection costs are constant. The structural steel and concrete costs for the boiler plant equipment are ~ $2/kW. The maximum change in these costs for the cases considered is ~ $0.20/kW and will be significantly less in general. The cost was thus assumed constant for this analysis. Erection cost changes are negligible.
4. Coal and stone feed size are assumed constant—1/4 in. x 0. This parameter is important when considering particle carry-over, but insufficient information is available to permit a quantitative analysis.
5. The ash and dust handling system cost is assumed constant. This cost will be affected by the particulate carry-over, but it is considered a second order effect for the cases considered.
6. Stack and foundation, instruments and controls, and other costs are constant.
7. All variables not being evaluated are assumed constant unless stated otherwise.

Table 1. PRESSURIZED FLUIDIZED BED BOILER POWER PLANT OPERATING AND DESIGN CONDITIONS

Cycle	
Steam system	2400 psia, 1000°F superheat, 1000°F reheat
Gas turbine expander	
Pressure ratio	10:l
Inlet temperature	1600°F
Air cooling	5%
Coal feed rate	53,910 lb/hr/module for nominal 300-MW plant design
Number of boiler modules	4
Boiler modules/gas turbine	2
Fuel/air ratio	0.0919
Boiler design	
Bed area	35 ft^2 (5x7 ft) – for ~80-MW module
Heat transfer surface	
Walls	2-in. OD tubes on 3-1/2 in. welded wall spacing
Bed	1-1/2-in. OD tubes in pre-evaporator and superheater; 2-in. OD in reheater (details in text)
Gas side heat transfer coefficient	50 Btu/hr-ft^2-°F
Tube materials	SA-210-A1 – pre-evaporator SA-213-T2 – lower superheater SA-213 T22 – water walls; upper superheater (lower loops); reheater SA-213-TP304H – upper superheater (upper loops)
Bed depth (expanded)	11 to 14 ft
Gas temperature drop from primary beds to gas turbine expander	150°F
Boiler operating conditions	
Bed temperature (100% load)	1750°F
Fluidizing velocity	6 to 9 ft/sec
Excess air	17.5%
Particle carry-over	~7 gr/scf
carbon from primary beds	6% of carbon feed
Auxiliaries	
Coal feed system	Petrocarb feed system
Primary particulate removal	4 size 355 VM 8/0/150 Duclone per module – nominal 300-MW design
Secondary particulate removal	2 model 18000 Type S collectors per module – nominal 300-MW design (quoted by Aerodyne Dev. Corp.)
Stack gas coolers	Conventional heat exchanger design.

Table 2. COST OF A 318-MW PRESSURIZED FLUID-BED BOILER

Pressure parts[a]	$1,777,000
Shell	935,000
Subcontracted and contracted equipment	435,000
Drafting	185,000
Home office	685,000
Sub-total	$4,017,000[b]
Erection	500,000
TOTAL	$4,517,000

[a] Pressure parts include tubing cost, headers, downcomers, risers, tube bending, tube welding, and water wall fabrication.

[b] Field erected ($3,856,000 shop assembled).

Table 3. PRESSURIZED FLUIDIZED-BED BOILER POWER PLANT COSTS

Equipment Costs	
Component	**Cost, $/kW**
Boiler plant equipment	
Boiler	14.49
Particulate removal	12.76
Piping/ducts	4.43
Stack and foundation	0.47
Coal handling and feeding equipment	14.94
Ash and dust handling system	1.55
Instruments and controls	3.10
Miscellaneous equipment	0.94
Steam turbine—generator equipment	44.14
Gas turbine—generator equipment	14.80
Other: land, structures, electric plant equipment, miscellaneous plant equipment, undistributed costs	70.38
Subtotal	182.00
Total capital cost (inc. escalation, IDC etc.)	265.00

($340/kW for conventional plant with wet scrubbing on same basis)

Energy costs	**mills/kWhr**
Fixed charges	6.44
Fuel	4.04
Dolomite	0.52
Operating and maintenance	0.71
	11.71

(13.45 for conventional plant)

BOILER PLANT EQUIPMENT

Operating Conditions

Bed Temperature

The full load design temperature is 1750°F. Lowering the design bed temperature increases the total amount of heat transferred in the bed and thus increases the steam turbine power generation. At the same time, lowering bed temperature decreases the gas turbine inlet temperature—assuming no burning above the bed—and thus decreases the total gas turbine power generation. The decrease in gas turbine power is larger than the increase in steam power, resulting in an overall decrease in plant power (Figure 4). Lowering the bed temperature increases the total amount of heat transferred in the bed and thus requires more heat transfer surface. Assuming the cross sectional area of the fluid bed (5 x 7 ft for a 300-MW nominal plant size) and tube size/tube pitch are constant, the expanded bed depth for each functional bed increases with decrease in bed temperature as shown in Figure 5. The bed depth of the two superheater beds is assumed to be the same for convenience. This will not affect the total heat transfer surface requirement and the resultant module height shown in Figure 6. The bed depth and module height can be reduced by enlarging the bed area and module diameter. However, since the module diameter of 12 feet is considered to be the largest shippable railroad size, increase in module diameter to accommodate additional heat transfer surface may not be economic for a 300-MW plant.

The effect of changing design bed temperature on the steam generator cost is calculated based on the boiler cost estimation shown in Table 2 and on the assumptions that the module diameter is constant at 12 feet and that the total number of modules is four based on turndown consideration. The cost (not including erection) of the 4-module steam generator as a function of bed temperature with constant module diameter (12ft) is shown as curves Ia and Ib in Figure 7. Curve Ia assumes the maximum allowable bed depth to be 20 feet. That means any bed with expanded bed depth larger than 20 feet will have to be split into two beds with their separate air plenums and freeboards. Curve Ib assumes that there is no restriction on maximum bed depth. The choice of 20 feet as the maximum allowable bed depth is arbitrary, just to show the importance of this variable on the cost of a steam generator. It is doubtful that the bed depth of each fluid bed can be unrestricted without creating undesirable bubble formation and slugging, poor bedtube heat transfer coefficient, and temperature gradients in the bed at some bed depth. The maximum allowable bed depth at specific operating conditions will have to be experimentally determined in a large unit. Without the required experimental evidence, the steam generator cost (not including erection) is plotted against the maximum allowable bed depth in Figure 8. The bed temperatures were calculated by assuming the gas turbine temperatures of 1600, 1500, 1400, and 1300°F and by assuming a linear temperature loss between the boiler and the gas turbine inlet. The cost of the steam generator designed for 1636°F increases ~20 percent (~ $2.8/kW) over that designed at 1750°F if the maximum allowable bed depth is 15 feet. The cost increase is primarily due to the splitting of beds with bed depth higher than 15 feet. Bed splitting can be avoided by either decreasing boiler tube diameter and spacing in the bed or increasing the module diameter. Decreasing tube diameter and spacing will change the bed-tube heat transfer coefficient, tube bending and fabrication, and tube wall thickness. Increasing the module diameter will not only change the cost of the pressure shell but also affect construction—complete shop-assemblage versus partial field erection. All these factors have to be taken into account in designing an optimal boiler. These factors are discussed in separate sections.

In the present cost estimation, the possibility of using thinner wall tubes for the designs at lower bed temperatures was also taken into account by calculating the minimum tube wall thickness requirement. No allowance for corrosion is provided.

Change in the operating bed temperature will also change the gas temperature to both the primary and secondary cyclones and thus change the actual volumetric gas flow rate. This will change gas inlet velocity to the cyclones which, in turn, affects cyclone collection efficiency. This effect was estimated to be small compared to the effect of the change in pressure drop across the bed due to a change in the design bed temperature. Decreasing the design bed temperature increases the heat transfer surface requirement in the bed, which requires an increase in pressure drop due to an increase in bed depth if bed area and boiler tube configuration are constant. The decrease in net plant power output as a function of the design bed temperatures is presented in curve 1, Figure 9. The effect is small—a decrease of only ~0.3 percent if the design bed temperature is reduced to 1407°F.

Bed temperature is one of the primary variables used for load turndown in the present design. A 4:1 turndown can be met if the design bed temperature is higher than 1600°F. The primary limitation on the operating bed temperature is the sulfur removal efficiency of the sorbents in the bed. At bed temperatures higher than 1750°F or lower than 1350°F, the sulfur removal efficiency in the bed is too low. Thus it is concluded from the above bed temperature analysis that the design bed temperature should be the highest temperature required for desirable degree of load turndown and sulfur removal in the bed.

Fluidizing Velocity

At constant fuel feed rate and excess air, increasing the fluidizing velocity requires a decrease in the bed area and in the module diameter (Figures 10 and 11). For a constant overall heat transfer coefficient and a specific design bed temperature the total heat transfer surface in the bed is constant; a decrease in the bed area will require an increase in the bed depth at constant tube size and tube spacing and thus an increase in the module height. An economic design will depend on the balance of thse factors.

The bed area and bed depth requirements with respect to change in fluidizing velocity at different design bed temperatures are calculated. The corresponding module height and module diameter are presented in Figure 11. The cost of the pressure shell at different inside diameters is estimated based on the data from Foster Wheeler Corporation[1] and on an independent estimation by Westinghouse (Figure 12). The discontinuity at a module inside diameter of 12 feet is due to the cost difference between the shop assembled and the field erected shell.

One additional cost which has to be taken into consideration is the fabrication cost. In addition to the shell cost, the change of fabrication cost of water walls and tube bending cost are not to be ignored. Taking into consideration the factors involved, the steam generator cost is plotted against the superficial fluidizing velocity in the pre-evaporator ash shown in Fugure 13 for 318-MW and 635-MW plant. The sulperficial fluidizing velocity in the pre-evaporator is used here since it is the largest velocity in all beds inside a single module. The superficial fluidizing velocity in the superheaters and reheater can be calculated accordingly. the results show that increasing the fluidizing velocity tends to increase rather than decrease the total steam generator cost at a 318-MW plant size. Decreasing the fluidizing velocity in the pre-evaporator below ~ 8 ft/sec requires a shift from the shop-assemblage to the field erection and escalates suddenly the steam generator cost. A minimum cost does exist for a 635-MW plant. Figure 13 is for unrestricted maximum allowable bed depth. If the maximum allowable bed depth is limited to say 10 or 20 feet, the disadvantage of increasing the fluidizing velocity would be even larger at 318-MW size. At 635-MW size, the minimum would shift to lower velocity.

The important thing here is to understand why increasing the fluidizing velocity increases the steam generator cost at 318-MW size and produces a minimum at 635-MW. To better illustrate the point, cost reduction due to decrease in module diameter and cost escalation due to increase in module height for a four-module design are shown in Figure 14 for the design bed temperature at 1750°F. Increasing the fluidizing velocity escalates the steam generator cost almost linearly from the basic design point due to increase in module height (curve 2). At the same time, the cost decreases due to decrease in module diameter; however, the decrease is much more gradual and levels off at higher fluidizing velocity (curve 1). This is because the bed area alone occupies less than 40 percent of the total cross-sectional area of a pressurized module. The remaining area is required for piping and headers; this space is relatively unchanged at a specific plant size even though the bed area is reduced to increase the fluidizing velocity. At 635-MW size, however, the cost reduction due to decrease in module diameter is larger during initial deviation from the basic design point and thus creates a minimum (Figure 14).

Another approach for analyzing the effect of fluidizing velocity would be to change the number of modules as well as the module diameter. It is preferred to have a 5-module design based on turndown consideration; however, if a 3-module design shows a substantial saving with negligible effect on turndown capability, it would be a better choice.

Change in fluidizing velocity will change the total bed area required for each functional bed, but the total bed volume for each functional bed will remain constant once the tube size and spacing are fixed. Thus a design with smaller number of modules will require a larger module diameter at the same design fluidizing velocity. An estimation can usually be performed to evaluate the relative economy between these two designs. For example, consider a 4-module and a 3-module design at the same design fluidizing velocity and with the same bed depths. Since the bed volume of each functional bed is constant at fixed tube size and spacing for both cases, we have

$$\frac{D_4}{D_3} = \frac{3}{4} \qquad (1)$$

if bed height is assumed constant for both cases and the bed area occupies a fixed percentage of the total cross-sectional area of a module. D_4 and D_3 are the respective module diameters for the 4-module and 3-module designs. Since the shell cost is dependent on the vessel diameter (Figure 12), the relative advantage of these two designs will depend on the plant size in question. For example, for $D_4=12.5$ feet, D_3 can be calculated from equation (1) to be 14.4 feet. The shell cost can be found from Figure 12 to be \$0.94 x 10^6 for the 4-module design and \$0.79 x 10^6 for the 3-module design. If fabrication cost of the module internals is similar in both cases, the 3-module design will have a slight economic advantage. However, this advantage becomes progressively smaller because of rapidly increasing shell cost at large module diameter and rapidly decreasing shop-fabricatable portion in the design. It is estimated that the largest module diameter which is still economic for the 3-module design is ~17 feet. This conclusion is based on the assumption that boiler turndown is not a problem. If the module diameter is in the shop-fabricatable and railroad-transportable range, i.e., < 12 feet, designing for the maximum shippable module will have definite advantages provided that turndown is not a problem.

It is concluded from this analysis that for a plant size around 300 MW, the module diameter should be the largest within shipping limitations (12 feet for railroad transportation); at a 600-MW plant size, an optimum fluidizing velocity exists, and it should be found for each capacity. However, the cost deviation from that of the optimum design is less than \$1.00/kW (Figure 13). Of course, decreasing the bed area may reduce the number of feed points but the saving is only ~ \$0.10/kW. Although the effect of the fluidizing velocity on the steam generator cost is primarily based on the basic design conditions, i.e., bed-tube heat transfer coefficient = 50 Btu/ft^2-hr-°F, the trends would be the same for bed-tube heat transfer coefficient at 35 or 75 Btu/ft^2-hr-°F. Change in tube size and spacing may change the slope of the curves or alter the minimum in Figure 13; the conclusions will remain the same.

The effect of changing fluidizing velocity on the bed-tube heat transfer coefficient, combustion efficiency, and total particulate carry-over is not taken into account in this analysis due to lack in accurate quantitative data. The effect of dust loading and particle size distribution on the cost of the particulate removal system is evaluated separately.

Excess Air

Change in design excess air will affect the cycle efficiency and the cost of the boiler module, the steam and gas turbine equipment, and the particulate removal system. In order to quantify the effect of excess air on total boiler cost, the air/fuel ratio is allowed to vary with the total fuel input kept constant. To simplify the analysis, other parameters—bed temperature, tube size and tube spacing, bed-tube heat transfer coefficient, fluidizing velocity, and number of boiler modules—are held constant at the basic design values.

Thus, when excess air is increased beyond the design value, bed area has to be increased if the fluidizing velocity is kept constant. When the bed area is increased, the module diameter has to be increased as well; however, the module height is decreased due to a decrease in bed depth. Increasing air input into the bed will also increase the amount of heat carried out from the bed by air and reduce the total heat transferred in the bed. At constant bed-tube heat transfer coefficient, the total heat transfer surface requirement is reduced. At constant tube size and tube spacing, the bed depth is also reduced as well as the total module height. At 100 percent excess air, a reduction of >40 percent in heat transfer surface and a reduction of >30 percent in module height is possible. The bed depths for different functional beds are reduced to $\sim$4 feet which decreases pressure drop through the beds and increases cycle efficiency. The module diameter, in turn, increases from the original 12 feet inside diameter to more than 16 feet for an 80-MW module. Transferring all these changes into economics, an increase in excess air can reduce the boiler cost up to $\sim$\$0.60/kW as shown in Figure 15. Cost reduction due to heat transfer surface increases continuously with respect to excess air because of the decrease in the total amount of heat transferred in the bed. Cost reduction due to pressure shell first increases because of reduction in module height and then decreases because of increase in module diameter.

Increasing excess air will increase the overall plant efficiency as shown in Table 4. Larger gas turbines or additional gas turbines are needed to handle the increased mass flow of gas. If additional units are used, the increase in gas turbine equipment cost is shown as a cost adder in Figure 16. This does not account for cost reductions which can be realized by going to larger turbine capacities. This cost increase in gas turbine equipment is partially offset by a decrease in steam turbine equipment cost also shown in Figure 16. The major equipment items taken into consideration in this analysis include gas turbines with external manifolds, steam turbine system, circulating water and condensing systems, feedwater

Table 4. EFFICIENCY CALCULATIONS AT VARIABLE AIR FLOW RATES

Excess air, %	Fuel/air ratio	Plant size, MW	Heat rate, Btu/kWhr	Gas turbine, MW	Steam, MW	Gas turbine/ steam ratio	No. of gas turbine
17.5	0.0861	644.1	9026	127.9	532.2	0.240	2.000
50.0	0.0674	648.0	8972	156.9	506.3	0.309	2.450
90.0	0.0532	649.7	8948	193.5	470.4	0.411	3.025

system including station piping, and stack gas coolers.

Instead of keeping the fluidizing velocity constant, the bed area and module diameter can be kept constant and allow the fluidizing velocity to increase with excess air. In this case, the cost reduction in heat transfer surface will be similar, but the cost reduction in the shell will continuously increase with increasing excess air and not go through a maximum (Figure 15). The cost reduction in heat transfer surface and pressure shell at 100 percent excess air in this case (with 10 to 15 ft/sec fluidizing velocity) is estimated to be ~$1.00/kW. However, increasing the fluidizing velocity to larger than 15 ft/sec may be impractical in this design approach.

Increasing the excess air will decrease the total heat transfer surface required in the fluid bed until no boiler tube surface will be required at an excess air of approximately 300 percent. In this case the power system would become a combined cycle plant with the gas to the turbine expanders supplied from a coal-fired, adiabatic combustor. The heat recovery boiler would probably be unfired. This system concept has several significant differences from a pressurized fluidized-bed boiler power plant concept: for example, the boiler becomes an adiabatic combustor, particulate removal equipment costs increase significantly due to the increased gas flow, gas piping costs increase and the gas turbine power contribution increases from ~20 percent up to ~70 percent. An economic analysis of this system has not been made as part of this evaluation. The heat rate for the adiabatic combustor plant is projected to be 100 to 500 Btu/kWhr (depending on the gas turbine inlet temperature) greater than for the pressurized boiler plant. Further evaluation of this high excess air case is required to perform a comprehensive assessment.

Increasing the excess air will provide more flexibility in turndown. At 100 percent excess air, an additional ~10 percent load reduction is possible as compared to operation at 10 percent excess air. This means a boiler designed at 1600°F and 100 percent excess air will be able to meet a 4:1 turndown requirement. An adiabatic combustor system should extend the turndown capabilities.

More discussion on excess air and gas flow rate appears in the section on particulate removal.

Operating Pressure

The full load design pressure is 10 atm. When the design pressure level is changed and the other operating parameters remain constant, the gas density will change in proportion to the pressure, and the volumetric flow will vary accordingly. Therefore, the bed cross-sectional area will have to be changed to maintain constant fluidizing velocity and the bed depth changed to maintain constant bed volume. The heat transfer coefficient may change because of changes in the quality of fluidization.

The changes in volumetric flow and in gas density will affect the design of the particulate removal equipment. Gas turbine cycle efficiency is also dependent on the operating pressure. However, analysis of the high pressure fluidized-bed boiler system indicates a reverse direction in pressure level effect. Auxiliary equipment such as the coal and dolomite feeding systems are pressure dependent as well. The relative importance of these factors with respect to operating pressure is analyzed in the following paragraphs.

First, consider the boiler module alone. At constant fuel feed rate and excess air, increasing the operating pressure will decrease the gas volumetric flow rate. There are two approaches in designing the boiler modules: (a) keep the bed area constant and let the fluidizing velocity change with the operating pressure, and (b) keep the fluidizing velocity constant and change the bed area according to the operating pressure. The incremental module cost for the constant bed area case is the cost of reinforcing the pressure shells. Since for a constant overall heat transfer coefficient and a specific design bed temperature, the total heat transfer surface in the bed is constant. This amounts to $0.20/kW and $0.30/kW for operating pressures of 15 and 20 atm, respectively, at 300-MW nominal plant size. At 600-MW plant size, the respective cost increments are $0.10/kW and $0.20/kW. The credit of decreasing particulate carry-over by operating pressure will require a decrease in bed area. Since the total heat transfer surface is constant, a decrease in bed area will require an increase in bed depth at constant tube size and tube spacing, and thus an increase in the module height. In this case, the effect of operating pressure on boiler design and boiler cost while keeping the fluidizing velocity constant is the same as the effect of changing fluidizing velocity with the operating pressure constant. Thus the Figures 10, 13, and 14 can also be applied for this approach if the coordinate for the fluidizing velocity is changed to (fluidizing velocity at basic design) x (new operating pressure/basic design pressure). For a design bed temperature of 1750°F, the cost increments are $0.80/kW and $1.90/kW for operating pressures of 15 and 20 atm, respectively, at 300-MW plant size. At 600-MW plant size, the cost increvents are $0.20/kW and $1.20/kW respectively.

Comparing these two design approaches, the constant area case is the less costly one. Moreover, if basic design bed area and fluidizing velocity are maintained, increasing operating pressure will mean a higher capacity shop-fabricatable module (i.e., module diameter <12 ft). At 15 atm operating pressure, a module of ~120-MW capacity can be shop-fabricated; at 20 atm pressure, the maximum shop-fabricable module is ~ 160 MW. However, the module height will be considerably increased because of increase in total heat transfer surface required. The total increase in module height will depend on the heat transfer surface arrangement in the bed.

Increasing the operating pressure reduces the size of the particulate removal equipment because of the decrease in volumetric flow rate. It reduces the particulate removal efficiency as well because of changes in gas density and viscosity. Increase in operating pressure will also require reinforcement of the containment vessel and the piping and ducting. The savings in the particulate removal equipment by operating at 15 and 20 atm are estimated to be $4.0/kW and $6.0/kW, respectively, for a 300-MW plant size. The major saving comes from the secondary collectors where maximum single unit capacity is assumed restricted to 30,000 acfm. An increase in operating pressure will result in fewer units. Cost reduction by operating at higher pressure will be even greater for larger plant sizes or at higher design excess air.

Cycle optimization calculation was performed to evaluate the effect of operating pressure. The parameters studied are: intercooled and non-intercooled compression; gas turbine compressor pressure ratios from 10 to 30; and cycle gas side pressure drops of 3 to 8 percent. The results are summarized in Figure 17 which gives plant heat rate for the intercooled and non-intercooled cases. For the non-intercooled case, the best efficiency is obtained at a pressure ratio of 10; for the intercooled case, the optimum pressure ratio is 15 but with a higher heat rate and a more complex gas turbine. Thus an increase in operating pressure higher than 10 atm decreases the overall plant efficiency. This decrease is small however: ~ 0.2 percent at 15 atm.

Weighing the above discussions, the only distinct advantage for operating at pressures higher than 10 atm is the capability of shop-fabricating a large capacity plant, especially at higher design excess air.

Boiler Design

Heat Transfer Surface

Configuration

In the 300-MW design, the heat transfer surface is provided by serpentine tubes having the horizontal sections spaced as shown in Figure 18. Tubes of 2-in. OD are used at waterwalls and are space 3-1/2 inches apart. Tubes for pre-evaporator and superheaters are 1-1/2-in. OD and tubes for the reheater are 2-in. OD. The tubes can usually be arranged in staggered or rotated diamond arrays, or they can be arranged in a square or rectangular pitch (Figure 19).

The effect of tube pitch/diameter ratio on the cost of the steam generator (not including erection) for constant 12-ft module diameter was evaluated for three different tube sizes and tube spacings with respect to change in the design bed temperatures. The results are plotted in Figure 7. Curve 2 represents the estimated cost for a staggered arrangement of 1-in. OD tubes, where H = 4 inches and V = 2 inches (see Figure 19 for definition) in all beds. Curves 3a and 3b are for staggered arrangement of 2-in. OD tubes where H = 8 inches and V = 4 inches in all beds. Some interesting trends are present when these results are compared to the steam generator cost for the basic design. Ignore curves 1b and 3b for the time being because the assumption of unrestricted maximum allowable bed depth is not considered reasonable. Then:

1. Decreasing tube size and tube spacing increases the steam generator cost at design bed temperatures above ~1520°F (curve 2). Further decrease in bed temperature necessitates splitting the reheater bed in the basic design into two beds and substantially increasing the steam generator cost of the basic design.
2. Increasing tube size and tube spacing also increases the steam generator cost (curve 3a).

The reasons for these results are as follows:

1. When tube size and tube spacing are decreased, more heat transfer surface can be immersed in a unit bed volume which results in lower bed height and module height; thinner wall tubes can be used which results in lower tubing cost. These are positive advantages.
2. However, smaller tube size and tube spacing increase the amount of tube bending and tube welding required. This is because more tubes of smaller diameter are required to carry the same water/steam load at a constant flow rate in the tube. Fabrication cost as a function of tube wall thickness is not taken into account because of not enough information available. Pumping costs, a part of total operating cost, are not included here.

The balance of these two factors determines the total steam generator cost.

Since the tubing cost constitutes only about 20 percent of the cost of the pressure parts, the increase in fabrication cost is a more important cost. This can best be illustrated in Figure 20 where the component costs are plotted against the available heat transfer surface per unit volume which relates to the tube pitch/diameter ratio. Shell cost increases almost linearly with decreasing heat transfer surface per unit bed volume. The cost of tubing, headers, downcomers, and risers is almost constant at heat transfer surface larger than 6 ft^2/ft^3 bed volume; it increases rapidly at heat transfer surface lower than ~6 ft^2/ft^3 bed volume, which corresponds to the use of a tube diameter of 1-1/2-in. OD or larger. When larger tubes are used, the minimum wall thickness increases rapidly and so does the tubing cost which contributes most of the cost escalation at lower heat transfer surface per unit bed volume. However, the cost of tube bending, tube welding, and water walls fabrication increases steadily with increase in heat transfer surface per unit bed volume. However, the cost of tube bending, tube welding, and water walls fabrication increases steadily with increase in heat transfer surface per unit bed volume. The balance of all these factors creates a minimum in total steam generator cost at about 6.5 ft^2 heat transfer surface per ft^3 of bed volume. This can be achieved by arranging 1-in. OD tubes where H = 4 inches and V = 3 inches. Fortunately, the tube size and tube spacing used in the basic design is very close to this actual minimum. Clearly, there are different minimums at different design bed temperatures. An optimum design for a specific operating condition requires a separate evaluation. However, this optimum design point is not as critical as may be generally conceived. For example, at a design bed temperature of 1750°F (Figure 20) the difference in the steam generator cost between the optimum design and other designs is within $1.00/kW for available heat transfer surface of 3 to 11 ft^2/ft^3 bed volume, which covers the three tube sizes and tube spacings in our current evaluation.

The maximum allowable bed depth is a more important variable. The steam generator costs are also plotted against the maximum allowable bed depth at constant design bed temperature and constant bed area in Figures 21 and 22. Although the steam generator of 1-in. OD tubes (where H = 4 inches and V = 2 inches) is not as economical compared to the basic design, it becomes progressively more attractive at a lower maximum allowable bed depth. At maximum bed depths lower than 14 feet (Figure 21), a steam generator using 1-in. OD tubes is actually cheaper than the basic design by up to ~ $3.1/kW. At a maximum allowable bed depth of 10 feet, a $3.1/kW saving is realizable by using 1-in. OD tubes over that of the basic design at a design bed temperature of 1750°F and a $3.3/kW at a design bed temperature of 1636°F.

In conclusion, although using smaller tubes and tube spacings will increase the amount of heat transfer surface immersed in a unit bed volume, the economy is not always favorable. The accompanying increase in cost for tube bending, tube welding, and fabrication sometimes overtakes the savings in tubing cost and module height. However, smaller tubes and tube spacings do show more advantages when the maximum allowable bed depth is limited. Moreover, there is a definite minimum bed depth when cost is plotted against the heat transfer surface per unit volume. This can be utilized to find the optimum tube size and tube spacings.

In some cases, the tube pitch/diameter ratio effect on bed-tube heat transfer coefficient has to be taken into account in evaluating the optimum tube size and tube spacings. Data indicate that smaller tube spacing tends to decrease the heat transfer coefficient[3,4,6] as shown in Figures 23 and 24. Lowering the pitch/diameter ratio from 8 to 2 results in an increase in the heat transfer surface per unit volume of bed by a factor of 16 and decreases the heat transfer coefficient by only 18 percent (Figure 23). Thus, the major consideration in determining the pitch/diameter ratio is not the heat transfer coefficient, but the compactness and the cost of the boiler design. However, there is a gap between tubes, depending on the particle size, below which a sharp drop of heat transfer coefficient will occur (Figure 24). Determination of this minimum gap requires further experimental studies. Although this conclusion is drawn from the experimental evidence at atmospheric pressure, it should be qualitatively valid at atmospheric pressure. Heat transfer coefficients for the tubes at different bed depths may also be different for the pressurized conditions; unfortunately there is no quantitative information available.

An added advantage of using smaller tube sizes and tube spacings is that lower bed depth tends to decrease the pressure drop through the fluid beds and thus to increase the gas turbine efficiency (curve 2, Figure 9).

Heat Transfer Coefficient

In the basic design, the overall heat transfer coefficients assumed are 47 Btu/ft^2-°F-hr for the pre-evaporator, 45 Btu/ft^2-°F-hr for the superheater, and 43 Btu/ft^2-°F-hr for the reheater. The bed-tube heat transfer coefficient is assumed to be 50 Btu/ft^2-°F-hr for all beds. When the bed-tube heat transfer coefficient is changed, the overall heat transfer coefficient will be changed as well. This leads to a change in the total heat transfer surface requirement and the bed depth.

Change in the heat transfer coefficient will also change the tube metal temperature. A change of tube material may be necessary for some cases. The design metal temperature is assumed to be the maximum outside tube wall temperature based on the minimum permissable wall thickness.

Taking into consideration the aforementioned factors, the total steam generator costs at bed-tube heat transfer coefficients of 35 and 75 Btu/ft^2-hr-°F were projected for different tube pitch/diameter ratios at different design bed temperatures. At a low bed-tube heat transfer coefficient (35 Btu/ft^2-hr-°F) where a large amount of in-bed heat transfer surface is required, a boiler with smaller tube size and tube spacing is much more economical than the one with larger tube size and tube spacing. Saving up to 20 percent of the total steam generator cost is feasible if bed area is constant and maximum allowable bed depth is 20 feet. If the maximum allowable bed depth is less than 20 feet, the saving will be even larger. At a bed-tube heat transfer coefficient of 75 Btu/ft^2-hr-°F where the heat transfer surface requirement is substantially reduced, smaller boiler tubes and spacings do not have a clear advantage. If the bed-tube heat transfer coefficient is 35 Btu/ft^2-hr-°F rather than 50 Btu/ft^2-hr-°F as assumed in the basic design, the steam generator cost will increase by \$2.7/kW at 1750°F design bed temperature and by \$5.4/kW at 1600°F. If 1-in. OD tubes are used where H = 4 inches and V = 2 inches, the cost escalation would be \$2.0/kW at 1750°F and \$3.4/kW at 1600°F. If the heat transfer coefficient is increased to 75 Btu/ft^2-hr-°F, the reduction in steam generator cost from that of the basic design is only marginal, ~ \$1.0/kW at 1750°F.

Figures 25 and 26 show the effect of the bed-tube heat transfer coefficient on the steam generator cost at constant bed temperature. An increase of the bed-tube heat transfer coefficient from 50 to 75 Btu/ft^2-hr-°F (a 50 percent increase) decreases the steam generator cost by ~ 10 percent (~ \$1.40/kW). A decrease of bed-tube heat transfer coefficient from 50 to 35 Btu/ft^2-hr-°F (a 30 percent decrease) increases the cost by ~20 percent (~ \$2.80/kW) (Figure 25). The curves start to level off at higher bed-tube heat transfer coefficients. Thus, further increase in bed-tube heat transfer coefficient larger than about 75 Btu/ft^2-hr-°F does not affect the cost substantially. However, further decrease in bed-tube heat transfer coefficient lower than 50 Btu/ft^2-hr-°F increases the steam generator cost rapidly, especially for large tube sizes and tube spacings and at lower design bed temperatures (Figures 25 and 26). A 40 percent increase in cost occurs when the bed-tube heat transfer coefficient decreases from 50 to 35 Btu/ft^2-hr-°F at design bed temperatures of 1636°F for 2-in. OD tubes where H = 8 inches and V = 4 inches (curve 3, Figure 26). It is recommended to design the steam generator at a bed-tube heat transfer coefficient about 75 Btu/ft^2-hr-°F if it is at all possible, and to avoid designing the steam generator at a bed-tube heat transfer coefficient lower than 50 Btu/ft^2-hr-°F, especially if large tubes and spacings are used and if lower bed temperatures are employed.

To complete the evaluation, the cost information was also prepared for other cases at different design and operating variables to show the interacting effect of tube pitch/diameter ratio, bed-tube heat transfer coefficient, maximum allowable bed depth, and the design bed temperature.

Again, the maximum allowable bed depth turns out to be the limitation of the steam generator design and cost, especially at low bed-tube heat transfer coefficient where large amount of heat transfer surface is required in the bed. In this case, a smaller tube size and tube spacing and a higher design bed temperature are preferred. At a maximum allowable bed depth of 10 feet, the boiler designed at 35 Btu/ft^2-hr-°F costs \$3.5/kW more than that designed at 50 Btu/ft^2-hr-°F and \$8.6/kW more than that designed at 75 Btu/ft^2-hr-°F at 1750°F design bed temperature. At design bed temperature of 1407°F, the figures are \$6.7/kW and \$12.5/kW, respectively. With 1-in. OD where H = 4 inches and V = 2 inches, the figures are \$3.3/kW and \$4.4/kW at 1750°F; and \$2.8/kW and \$8.1/kW at 1407°F, respectively.

Cost savings become smaller when the bed-tube heat transfer coefficient is further increased over 75 Btu/ft^2-hr-°F. If the bed-tube heat transfer coefficient is decreased to lower than 50 Btu/ft^2-hr-°F, the steam generator cost increases rapidly. Thus it is recommended that the bed-tube heat transfer coefficient be kept at higher than 50 Btu/ft^2-hr-°F and preferable around 75 Btu/ft^2-hr-°F at the current design conditions.

Tube Materials

The tube materials used in the basic design are conventional boiler tube material with SA-210-A1 for tubes in pre-evaporator; SA-213-T2 for tubes in lower superheater; SA-213-T22 for tubes in water walls, upper superheater (lower loops), and reheater; and SA-213-TP304H for tubes in upper superheater (upper loops). Changes in design bed temperature, bed-tube heat transfer coefficient or steam temperature may require higher grade tube materials; however, these changes do not substantially affect the steam generator cost because the tubing cost alone constitiutes only ~ 10 percent of the total steam generator cost (~ \$1.40/kW). Higher fabrication cost for higher alloy material may increase this cost slightly. Nevertheless, the total boiler cost is not expected to increase significantly due to change of tube material unless the operating bed temperature and heat transfer coefficient are drastically changed.

Module Capacity

Boiler modules can be shop fabricated, partially shop fabricated or field erected depending on the size. Modules up to 12-ft diameter can be shop fabricated. Modules up to 17-ft diameter can be partially shop fabricated—the boiler internals being shop fabricated, the pressure shell being field erected. The plant concept for a given capacity can be either multiples of shop fabricated modules, partially shop fabricated modules, or field erected modules.

The boiler plant equipment cost is different for each case. The steam generator cost will depend on operating conditions and design variables such as design bed temperature, tube size and tube spacing, maximum allowable bed depth, etc. Auxiliary equipment will also be affected: coal feeding, limestone or dolomite feed and withdrawal, particulate removal, steam piping, boiler feed water system, etc.

The evaluation of these approaches is based on Figure 3, which presents the cost variation of the pressure parts, . shell, subcontrated and contracted equipment, drafting and home office, and erection with respect to the plant size. Figure 27 presents the resulting costs (including erection) for design bed temperature at 1750°F; Figure 28 presents those for design bed temperature at 1636°F at a maximum allowable bed depth of 20 feet. The results show that at a plant size larger than ~ 340 MW, partially shop fabricated 4-module plants with maximum shop fabrication of the pressure parts are more economic than collective multiples of largest shop-fabricated modules of the same plant capacity. Changing the tube size and tube spacing does not affect this conclusion (Figures 27 and 28). Change in heat transfer coefficient should not produce a different conclusion because the cost escalation due to addition of a single module is more expansive than simple enlargement of the existing modules. However, this is no longer true when the shell size is larger than ~ 17 feet, because the degree of shop fabrication of the pressure parts decreases and the steam generator cost again increases at a much higher rate.

When the maximum allowable bed depth is decreased from 20 feet, the cost saving of the 4-module partially shop-fabricated plant is expected to increase. The splitting of the beds in the shop fabricated module means an increase in module height; however, in the partially shop-fabricated module the splitting of beds can be avoided by simply enlarging the module diameter. From the discussion on Figure 14, the latter means a more economic alternative except when the module diameter is increased beyond ~ 17 feet.

The optimum design variables (fluidizing velocity, excess air, and pressure) in relation to module capacity are discussed in their respective sections.

Particulate Removal System Economics

The primary variables taken into consideration in analyzing particulate removal system economics are dust loading, particle size distribution entering and leaving the system, and gas flow rate. The effects of boiler operating variables—bed temperature, freeboard height, and superficial velocity—and design variables—tube pitch/diameter ratio and bed-tube heat transfer coefficient—were also evaluated.

Range of Dust Loadings and Particle Size Distributions Considered

The cases evaluated are outlined in Table 5.

Gas Turbine Specification

A review of operating experience and assessment of erosion in gas turbines was prepared by Westinghouse under contract to the Office of Air Programs.[1] Specifications for the fluidized-bed combustion system based on that study are:

- Dust loading less than 0.15 gr/scf.
- Concentration of particles greater than 2 μm less than 0.01 gr/scf.

These design requirements will be updated as additional laboratory test data and operating experience become available.

Table 5. CASES EVALUATED FOR DETERMINING EFFECT OF DUST LOADING AND PARTICLE SIZE DISTRIBUTION

Cases evaluated	Dust loading leaving FBC	Particle size distribution	Cyclone system design
Group			
Case 1	Basic design (6.7 gr/scf)	(Refer to Figure 29)	Figure 30
Case 2	Double design value	Curve 1 for particles elutriated from FBC	Figure 30
Case 3	Triple design value	Curve 2 for particles elutriated from CBC	Figure 30
Case 4	Triple design value		Figure 31
Case 5	Triple design value		Figure 32
Group 2			
Case 1	Basic design (6.7 gr/scf)	Curve 2 for particles elutriated from FBC	Figure 30
Case 2	Double design value	Curve 3 for particles elutriated from CBC	Figure 30
Case 3	Triple design value		Figure 30

Effect of Dust Loading and Particle Size Distribution on Particulate Removal Equipment Cost

The effect of dust loading and particle size distribution leaving the boiler on the particulates going to the gas turbine for the particle removal systems selected are shown in Table 6. The gas turbine specification is exceeded in several cases. In order to meet the specification, alternative particulate removal systems could be considered — granular bed filters, electrostatic precipitators, ceramic filters, etc.; additional mechanical collectors could be used in series; or boiler operation could be altered. The additional use of mechanical collectors is the approach used to evaluate additional costs. This approach was selected since the equipment is available and thus provides the best cost data, and because the effect of boiler operating conditions on particulate emission is difficult to project. Table 7 presents a summary of the economic implications.

Table 6. DUST LOADINGS LEAVING THE SECONDARY CYCLONE FOR DIFFERENT CASES EVALUATED

Cases Evaluated	Total dust loading leaving secondary cyclone, gr/scf	Dust loading for particles $>2\ \mu m$, gr/scf
Group 1		
Case 1	0.14	0.007
Case 2	0.27	0.014
Case 3	0.40	0.020
Case 4[a]	0.16	0.0006
Case 5	0.40	0.019
Group 2		
Case 1	0.27	0.013
Case 2	0.46	0.022
Case 3	0.67	0.032

[a] Assuming same fractional collection efficiency for the second secondary cyclone as that for the first one. This assumption is too optimistic.

If the gas turbine dust loading requirement is defined as < 0.01 gr/scf without reference to particle size, no centrifugal separator presently available can meet this requirement within reasonable cost in all cases discussed above. If this were the case, high temperature ceramic filters may have to be used. The possibility of this application is being evaluated. In this respect, the particle size distribution is a far more important parameter than the dust loading, since the collection efficiency for the particle size smaller than 2 μm decreases rapidly. Thus, the particle size distribution curves assumed for the present evaluation (as shown in Figure 29) are conservative because a large amount of fines is assumed to be present.

Effect of Total Gas Flow Rate on Particulate Removal Equipment Cost

The effect of increasing gas flow rate was evaluated for four cases which correspond to the basic design flow rate, 30, 50, and 100 percent air. The selection of first stage cyclones is based on the criterion of maximum efficiency at minimum cost with a minimum cyclone efficiency of 85 percent. The cost increment for higher gas flow rate is shown in Figure 33.

The first stage cyclone cost includes not only the cost of the first stage separators supplied by Ducon but also the cost of the separator pressure vessel and all of the gas piping from the steam generator outlet to the secondary separator inlets. The costs for pressure vessel and gas piping were estimated for two cases. In one case, the gas piping from the steam generator to the first stage separator is lined with hard refractory, but the pressure vessel and the gas piping from it are lined with stainless steel. In the other case, hard refractory without an alloy liner was used throughout. At 100 percent excess air, the increase is \$1.70/kW and \$2.40/kW for hard refractory liner and stainless steel liner respectively. The major cost increase is from enlargement of pressure vessels and gas piping due to higher gas flow rate. The increase in separator cost alone constitutes only about 18 percent of the total cost increment.

Table 7. PARTICULATE REMOVAL SYSTEM COST

Case	Need for further clean-up to achieve gas turbine specification	Particulate removal system cost increase, $/kW
Group 1		
Case 1	No	-
Case 2	May not be required[a]	-
Case 3	Yes, a second secondary cyclone in series or granular bed filter	6.30 8.50
Case 4	No	-
Case 5	Yes, a second secondary cyclone in series or granular bed filter	6.30 8.50
Group 2		
Case 1	May not be required[a]	-
Case 2	Yes, a second secondary cyclone in series or granular bed filter	6.30 8.50
Case 3	Yes, a second secondary cyclone in series or granular bed filter	6.30 8.50

[a] Dust loading is larger than specification but particle size is close to specification.

The cost increase for the second stage is presented in Figure 34. The incremental cost for the second stage is more than 2 times that of the first stage ($5.00/kW versus $2.40/kW at 100 percent excess air). This is because model 18,000 is the largest cyclone now supplied by Aerodyne. The capacity of model 18,000 with dirty gas as the secondary gas is 30,000 ft^3/min. Any gas flow rate higher than that will require multiple units with their individual pressure vessel. This tends to increase the incremental cost of the second stage; however, the rate of cost increase slows down at excess air larger than 100 percent. At excess air larger than 100 percent, the rate of cost increase for the first stage speeds up. This is because increases in pressure vessel size and gas piping diameter increase the incremental cost rapidly at very high gas flow rate.

Taking into account the cost of gas piping from the second stage cyclones to gas turbine, the total incremental cost at different gas flow rate is shown in Figure 35.

Combining cost figures from Figures 15, 16, and 35, an increase in total boiler cost of $9.6 to $10.2/kW is required to operate the boiler at 100 percent excess air. This, however, does not take into consideration that the combustion efficiency in the primary beds will approach 100 percent at 100 percent excess air and that the carbon burn-up cell can be eliminated. In addition, the particulate removal system would be much simpler and the high excess air may also provide the necessary flexibility to achieve plant turndown if the boiler is designed at lower bed temperatures. An additional ~10 percent turndown capability is obtained by operating at 100 percent excess air. The lower bed depths would also permit combining the two superheater beds into one bed which would also reduce module height and cost. Thus it is concluded that if the carbon burn-up cell can be eliminated, operating at 100 percent excess air may result in a lower energy cost with increased cycle efficiency and flexibility. This conclusion is true at 300-MW nominal plant size, but is not necessarily true at 600-MW plant size. This is because at 600-MW plant size, the module has a 17-ft diameter at the basic design. Further increase in excess air requires either an increase in module diameter or in the total number of modules. At 100 percent excess air, four modules with 23-ft diameter are required. Since fabrication cost of the internals increases rapidly at module diameters larger than 17 feet because of rapidly decreasing shop-fabricable portion, an increase of $5.0/kW in boiler module cost alone is conceivable. Adding the cost increase in boiler cost is about $15.0/kW. In this case, operating at higher pressures may be beneficial.

Instead of increasing the module diameter from 17 to 23 feet, the number of modules could be increased keeping the module diameter constant. At 100 percent excess air, seven modules are required. In this case, in addition to module cost individual particle removal equipment has to be provided for each module; ducting and piping manifolds have to be increased; coal feeding systems become more complicated; and above all, instrumentation and control have to be more sophisticated. The total increase in boiler cost is estimated to be $20.0 to $25.0/kW in this case. Consequently, even if the carbon burn-up cell could be eliminated, the increase in cost and complication in control do not clearly favor the operation at high excess air for large plant size. A careful evaluation of overall design and control philosophy should be done if operation at high excess air is to be attemped for large plant size.

POWER GENERATION EQUIPMENT

Alternative boiler design and operating conditions will have three primary affects on the power generation equipment:

1. Capacity of gas and steam turbine equipment.
2. Gas turbine inlet temperature.
3. Ability to achieve higher steam temperatures and pressures.

Since this analysis assumes a constant fuel rate, the capacity changes are considered small except for the high excess air case. The effect of capacity was considered with the excess air analysis.

Gas Turbine Inlet Temperatures

The design value for the gas turbine inlet temperature was 1600°F for the base design of the pressurized fluid-bed boiler, which is well below the current state-of-the-art temperatures of 1800-1900°F for utility intermediate load applications. The 1600°F level was established by assuming the flue gas leaves the bed at the 1750°F bed temperature, and that the temperature difference between the bed and the gas turbine inlet would be 150°F. Several factors may alter the gas turbine inlet temperature. These include:

1. Temperature drop between boiler and turbine expander—150°F was assumed which is probably excessive. A drop as low as 50 to 75°F may be achieved.
2. Bed temperature—if the boiler design temperature is changed, the turbine inlet temperature will change. Sulfur removal considerations and ash agglomeration will determine the maximum temperature.
3. Combustion above the bed—combustion has been observed above the bed of a fluidized-bed boiler which increases the gas temperature 200-300°F. Any combustion above the bed would increase the gas turbine inlet temperature. No combustion was assumed in the base design.
4. Modification of the cycle to provide for reheat of the product gas from the boiler prior to the gas turbine. One concept for doing this is shown in Figure 36. Carbon carried out of the primary beds would be gasified to produce a low-Btu gas. The gas would be used in the second stage combustor.

Performance calculations were made to determine the effect of a change in the gas turbine inlet temperature on plant performance. The results are summarized in Table 8. These results are for 17.5 percent excess air and a boiler efficiency of 88.6 percent. A 200°F change in the turbine inlet temperature will change the plant heat rate ~1 percent using current technology.

Table 8. PERFORMANCE OF PRESSURIZED FLUID-BED BOILER POWER PLANT AS A FUNCTION OF GAS TURBINE INLET TEMPERATURE

Gas turbine inlet temperature, °F	Plant output,[a] MW	Plant heat rate,[a] Btu/kWhr	Fuel burned after primary beds,[b] %
1400	625.6	9293	-
1500	634.9	9157	-
1600	644.1	9026	-
1700	644.8	8921	2.5
1800	645.3	8820	5.0
1900	641.8	8773	7.6
2000	636.2	8753	10.1

[a] The decrease in performance at high gas turbine inlet temperatures is the result of increased bleed air required for turbine cooling and the increase in gas turbine waste heat which reduces steam cycle extraction for regenerative heating. The heat rate at 2000°F would be reduced ~270 Btu/kWhr if turbine blade cooling was not required.

[b] Assumes any increase above the design value of 1600°F has to result from burning fuel either above the bed or separately.

Steam Temperature

Plant performance can be increased by increasing steam temperature and pressure. The effect of higher steam temperatures on the performance of the plant is shown in Table 9.

Table 9. EFFECT OF HIGHER STEAM TEMPERATURES ON PLANT PERFORMANCE

Steam temperature, °F	Steam pressure, psi	Gas turbine inlet temperature, °F	Power, MW	Heat rate, Btu/kWhr
1000/1000	2400	1600	644.1	9026
1100/1100	3300	1600	673.9	8627
1200/1200	4500	1600	690.8	8417

An increase of 100°F in both superheat and reheat temperatures will give a reduction of about 400 Btu/kWhr in plant heat rate. The increased performance and inherently less severe boiler tube corrosion in fluidized-bed boilers make high steam temperatures attractive.

ASSESSMENT

A summary of the sensitivity analysis is presented in Table 10. Each parameter is indicated with a projection of what change might be required as the result of experimental data. For example, the bed temperature was set at 1750°F for 100 percent load. This temperature may prove to be too high for economic sulfur removal and have to be reduced to 1600 or 1650°F which may be more favorable. The summary table indicates how such a change would affect plant cost and performance assuming no other variable restrictions. In this case, the plant cost would increase < \$3/kW, plant performance would decrease < 0.5 percent, and plant turndown to 25 percent load could still be achieved. This would result in an energy cost reduction of < 0.3 mills/kWhr. Since the projected advantage over a conventional plant with stack gas scrubbing is greater than 1.5 mills/kWhr,[1] the penalty for lowering the temperature is not significant. This conclusion holds for all of the variables considered. The conclusion is generally valid for plant capacities of 200 to 700 MW. The results of the sensitivity analysis indicate that the base plant design is relatively insensitive to changes in operating conditions or design parameters.

Operating conditions and design parameter changes can occur which would result in a significant cost increase for the system. This would most likely occur as the result of additive problems. For example, suppose the bed temperature had to be decreased to 1600°F, the heat transfer coefficient was only 35 Btu/hr-ft^2-°F, and the dust loading from the boiler was 3 times the design value. The plant cost could increase by 7 to 8 percent and the efficiency decrease by

Table 10. SENSITIVITY ANALYSIS SUMMARY

Parameter	Change	Effect			
		Plant cost			
		Boiler	Auxiliaries	Power generation equipment	Performance
Boiler operating conditions					
Bed temperature	Reduction from 1750°F to 1600°F	< $3/kW[a] increase	Negligible	Negligible	≤ 0.5% decrease in efficiency 4:1 turndown requirement can be met
Fluidizing velocity	Decrease to 5 ft/sec	< $0.50/kW increase	b	—	—
	Increase to 15 ft/sec	∼$1/kW increase[c]	b	—	—
Excess air	Increase to 100%	< $1/kW decrease[d]	∼$7.50/kW increase	< $4/kW increase[e]	∼ 0.5% increase in plant efficiency Improved turndown capability
Pressure	Increase to 15 atm	< $0.50 kW increase for constant area ∼$1/kW increase for constant velocity	$4 /kW decrease	Negligible	∼ 0.2% decrease in plant efficiency
Particulate carry-over					
Loading	Increase loading to 3 times the design value (∼20 gr/scf)	—	$6-8/kW increase[f]	—	Negligible
Particle size	Increase fines: particles <10 µm increased from 15% to 25%	—	No increase projected[f]	—	Negligible
Boiler design					
Heat transfer surface					
Configuration	Increase available heat transfer surface per unit volume from 5.5 ft²/ft³ to 11 ft²/ft³	∼$1/kW increase[g]	—	—	—

Table 10 (continued). SENSITIVITY ANALYSIS SUMMARY

Parameter	Change	Effect			
		Plant cost			
		Boiler	Auxiliaries	Power generation equipment	Performance
	Decrease from 5.5 ft^2/ft^3 to 3 ft^2/ft^3	~ \$1/kW increase [g]	—	—	—
Heat transfer coefficient	Increase from 50 to 75 Btu/hr-ft^2 °F	~ \$1/kW decrease	—	—	—
	Decrease from 50 to 35 Btu/hr-ft^2 °F	~ \$3/kW increase	—	—	—
Materials	Assume tubing cost 50% greater than base design	< \$2/kW increase [h]	—	—	—
Bed depth [i]	Reduced to 10 ft	~ \$3/kW increase	—	—	—
Power generation equipment [k]					
Gas turbine inlet temperature	± 200°F from 1600°F	— [j]	— [j]	— [j]	± 1% in efficiency
Steam temperature	100°F increase in superheat and reheat	< \$2/kW increase	—	Negligible	~ 2% increase in efficiency

[a] Increase will depend on bed depth restrictions; \$3/kW would correspond to a maximum allowable height of ~15 ft.

[b] The fluidizing velocity will affect the particulate removal equipment – see particulate emission parameter for costs.

[c] Considerable savings may be realized for large capacity (>600 MW) plants since higher velocities avoid the need for field erection.

[d] Additional savings would be realized for large capacity (>600 MW) plants in order to avoid field erection. The \$1/kW does not include a cost reduction which may result from elimination of the carbon burn up bed due to increased efficiency.

[e] \$4/kW assumes the larger capacity gas turbine has the same unit cost as the base machine. Actual cost \$1/kW of a larger machine would be lower.

[f] Based on projected gas turbine requirement of <0.01 gr/scf of particles <2 μm.

[g] Assumes maximum allowable bed depth of 20 ft.

[h] Includes effect on fabrication.

[i] Assumes constant freeboard. (Change in freeboard requirement would have similar effect.)

[j] If the temperature is the result of equipment modification, the capital cost would be altered. Gas piping will be affected in any case.

[k] The plant cost projections are based on the base case plant capacity and performance. An increase in efficiency will reduce the specific plant cost. Water cooling has not been included. A higher efficiency will result in lower cooling equipment cost.

~0.5 percent. This would result in an increase in the energy cost of ~0.7 mills/kWhr. This is a significant increase but still within the economic margin. Caution must be exercised in interpreting multiple changes in the variables. In the case above, the cost effects were added. However, decreasing the bed temperature and increasing the heat transfer coefficient both increase the heat transfer surface and the bed depth if the bed area is maintained constant. Any restrictions on bed depth would also have to be considered in the evaluation. Parametric curves have been prepared to enable this type of evaluation to be made.

The effect of boiler plant pressure drop and the steam turbine condenser pressure on plant performance has been presented.[2] The boiler plant pressure drop has a small effect on plant capacity and heat rate: 1 percent increase in pressure drop results in a 0.1 percent increase in plant heat rate. An increase in the steam turbine condenser pressure from 1-1/2-in. Hg to 3-in. Hg for a cooling tower results in a 2.5 percent increase in heat rate.

The potential performance of the plant was evaluated by assessing the effect of higher gas turbine inlet temperatures and higher steam temperatures and pressures. The results indicate that plant efficiencies of ~45 percent can be achieved with gas turbine inlet temperatures greater than 2000°F, with high temperature blade material to minimize cooling requirements, and with steam temperatures of 1200°F.

Advances in boiler plant subsystem concepts have not been considered in this analysis. Cost reductions may be achieved by using alternative concepts. The following components have been studied for potential savings:

> Particulate Removal—The projected system utilizes four secondary collectors for final particle removal before the gas turbine. Alternative systems are being considered which may reduce the number and size of the units. Cost estimates indicate that reductions of $1 to $5/kW for the total particulate removal system may be possible.
>
> High-Temperature Gas Piping—The high-temperature gas piping cost would be reduced if the particulate removal system were simplified by using fewer units per module. Additional savings might be realized if refractory lined pipe could be used between the secondary collectors and the gas turbine. The present design uses a high-alloy steel to assure protection of the gas turbine from additional particulates.
>
> Coal Feeding System—The coal feeding system design is based on systems which have been built and operated. The design provides a separate coal feeding system for each fluidized bed in order to assure control of the coal feed rate to each bed. It may be possible, however, to reduce the number of coal feed systems from 16 to 4 if independent control of solids flow to each bed in a module can be achieved from a single pressurized injector. The potential cost reduction is estimated to be > $2/kW.
>
> Stack Gas Cooler Design—Cost estimates were obtained for the stack gas coolers, but no attempt was made to optimize the design or consider nonconventional designs, such as those using fluidized beds. Preliminary conceptual evaluation indicates that the cost might be reduced $4.40 to $3.40/kW.

CONCLUSIONS

1. Base plant design is near optimal.

2. Pressurized fluidized-bed boiler power plant maintains greater than 5 to 10 percent energy cost advantage over conventional plant with stack gas scrubbing.

> Effect of a potential change in bed temperature, fluidizing velocity, heat transfer surface configuration, gas side heat transfer coefficient, boiler tube

materials, bed depth limitations or pressure will result in: no significant change in plant operability or performance, < 2 percent increase in plant cost, and < 0.2 mills/kWhr increase in energy cost. Effect of increasing the dust loading to three times the design value will increase the plant cost ~ 4 percent, energy cost < 0.3 mills/kWhr.

Effect of increasing excess air will result in: increased turndown capability and performance, 3 to 6 percent increase in plant cost, and no significant change in energy cost.

3. Plant efficiencies of ~ 45 percent may be achieved for gas turbine inlet temperatures above 2000°F and steam temperatures of 1200°F.

ACKNOWLEDGMENT

This work was performed under contract to the Environmental Protection Agency, Office of Research and Monitoring. P. P. Turner served as contract officer. N. E. Weeks, Westinghouse Power Generation Systems Division, performed power system cycle analyses.

REFERENCES

1. Evaluation of the Fluidized Bed Combustion Process. Volumes I-III. Westinghouse Research Laboratories, Pittsburgh, Pa. Submitted to the Office of Air Programs, Environmental Protection Agency, Research Triangle Park, N. C. November 1971.
2. Keairns, D. L., J. R. Hamm, and D. H. Archer. (Presented at Annual AIChE Meeting. San Francisco. November 1971. AIChE Symposium Series, Volume "Air", 1972.
3. Highley, J., D. Chandrasekeva, and D. F. Williams. Fluidized Combustion Section, Coal Research Establishment, National Coal Board, London, England. Report Number 20, April 1969.
4. McLaren, J. and D. F. Williams. J. of Institute of Fuel. 303, August 1969.
5. Smith, S. Private communication, Boiler Tube Division, The Babcock & Wilcox Co., Barberton, Ohio.
6. Genetti, J. W. and W. E. Bartel. (Presented at American Institute of Chemical Engineers 72nd National Meeting. St. Louis. May 22-24, 1972.)

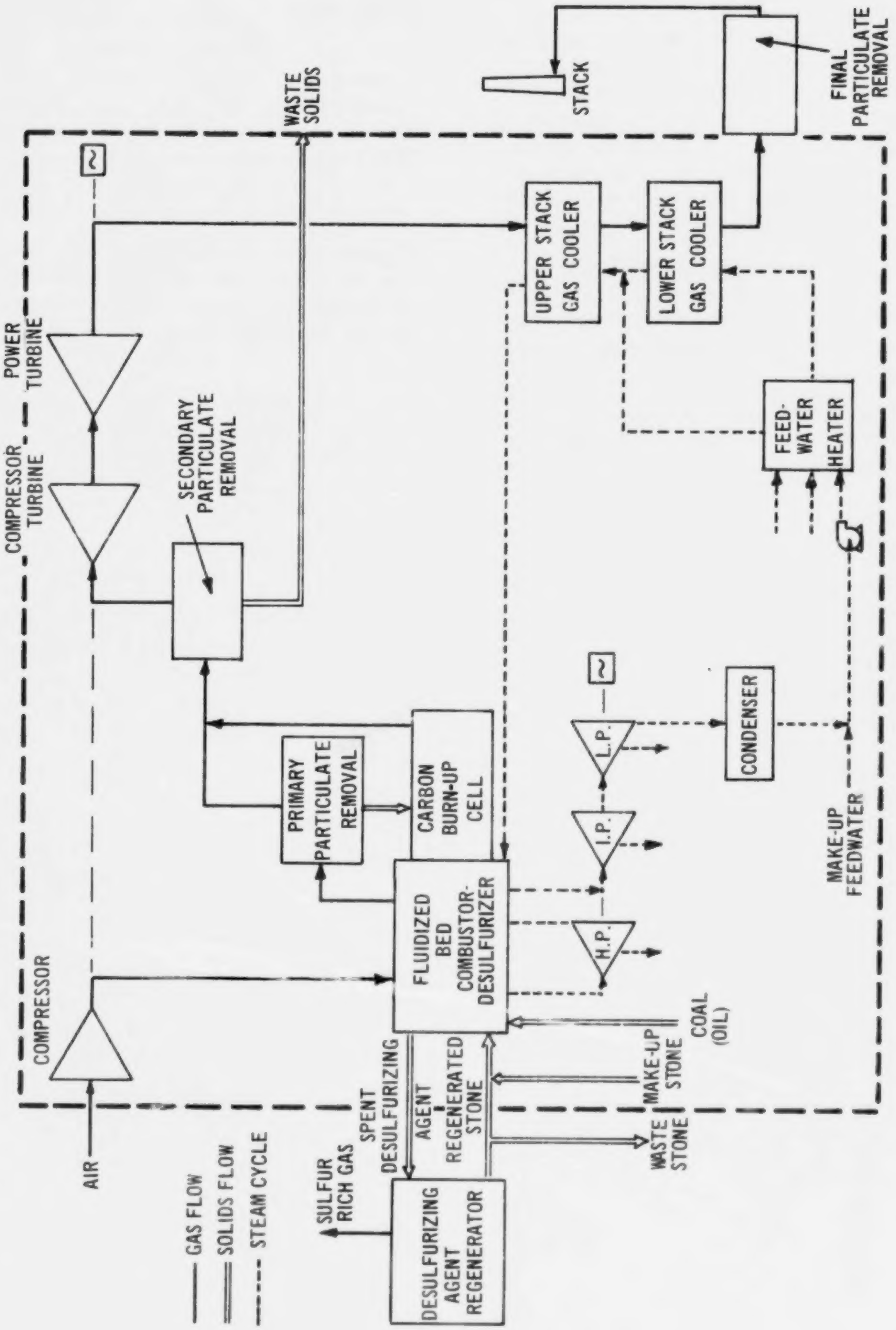
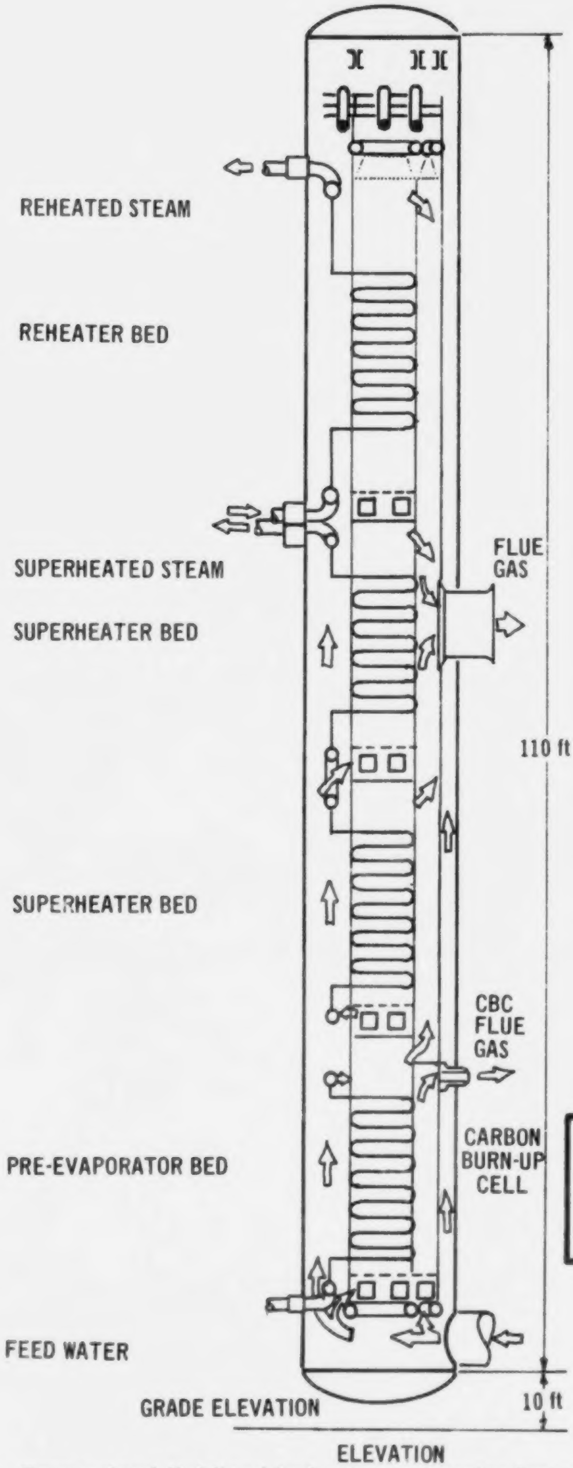


Figure 1. Pressurized fluid-bed boiler power system.



PLANT SIZE	VESSEL DIAMETER
320 MW	12 ft
635 MW	17 ft

Figure 2. Pressurized fluidized-bed steam generator for combined cycle plant (four required).

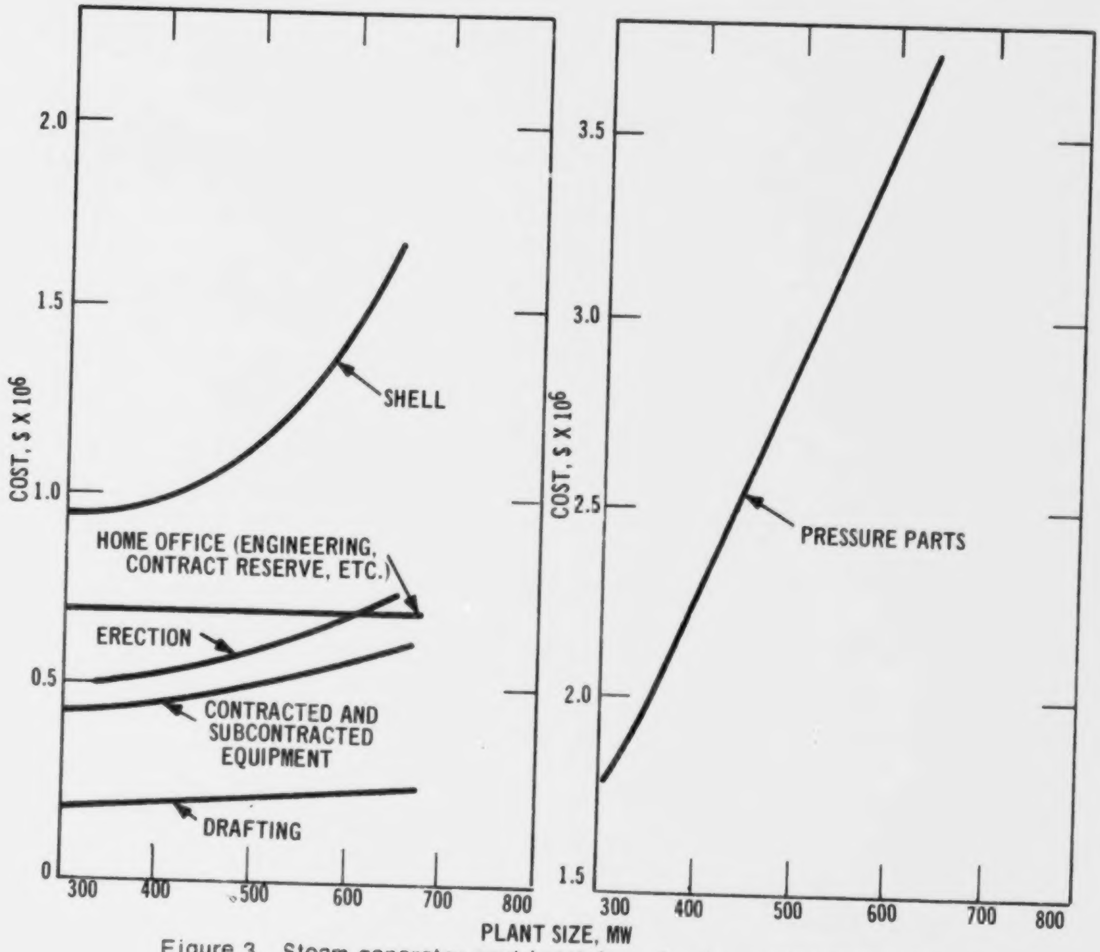


Figure 3. Steam generator cost breakdown for the W-FW basic design.

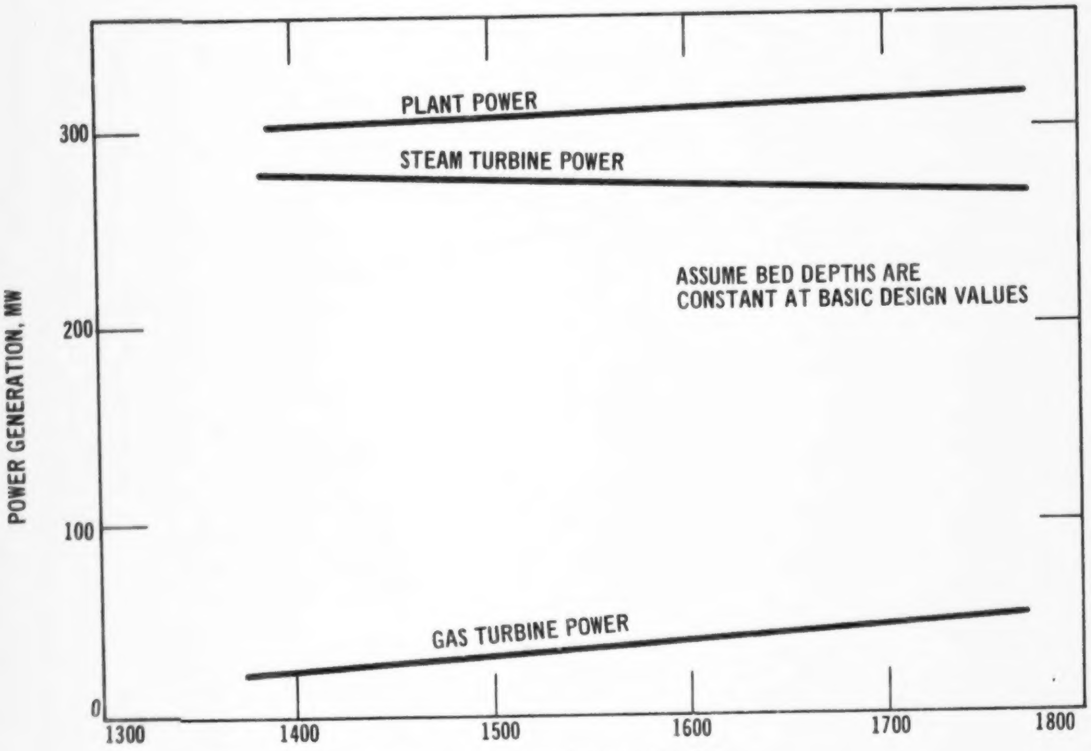


Figure 4. Effect of bed temperature on power generation.

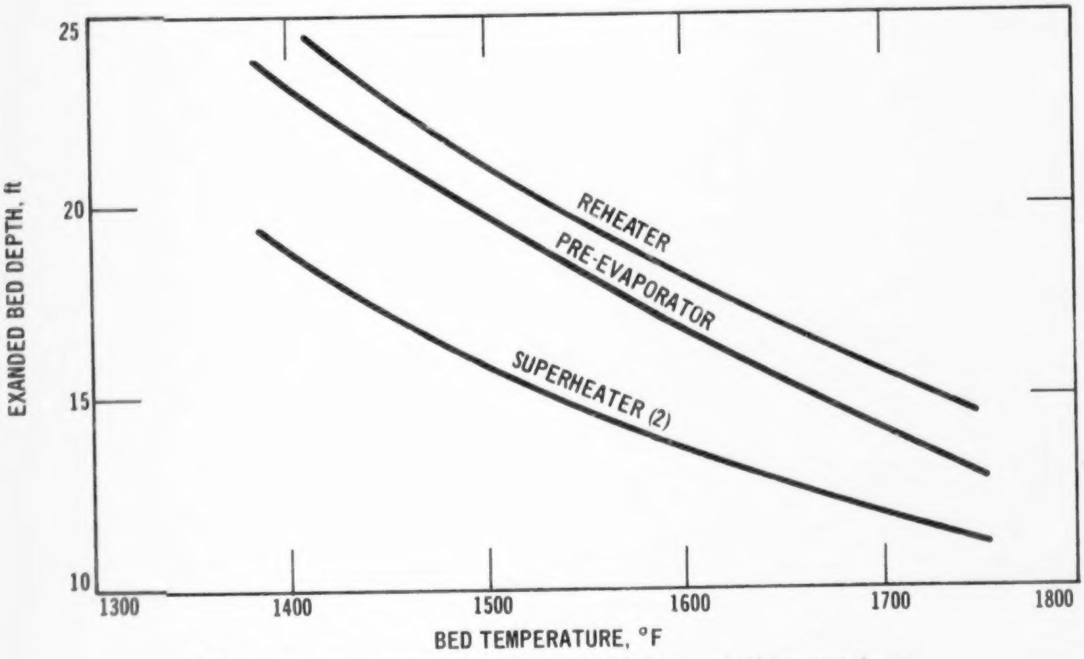


Figure 5. Bed depth requirement at different bed temperatures.

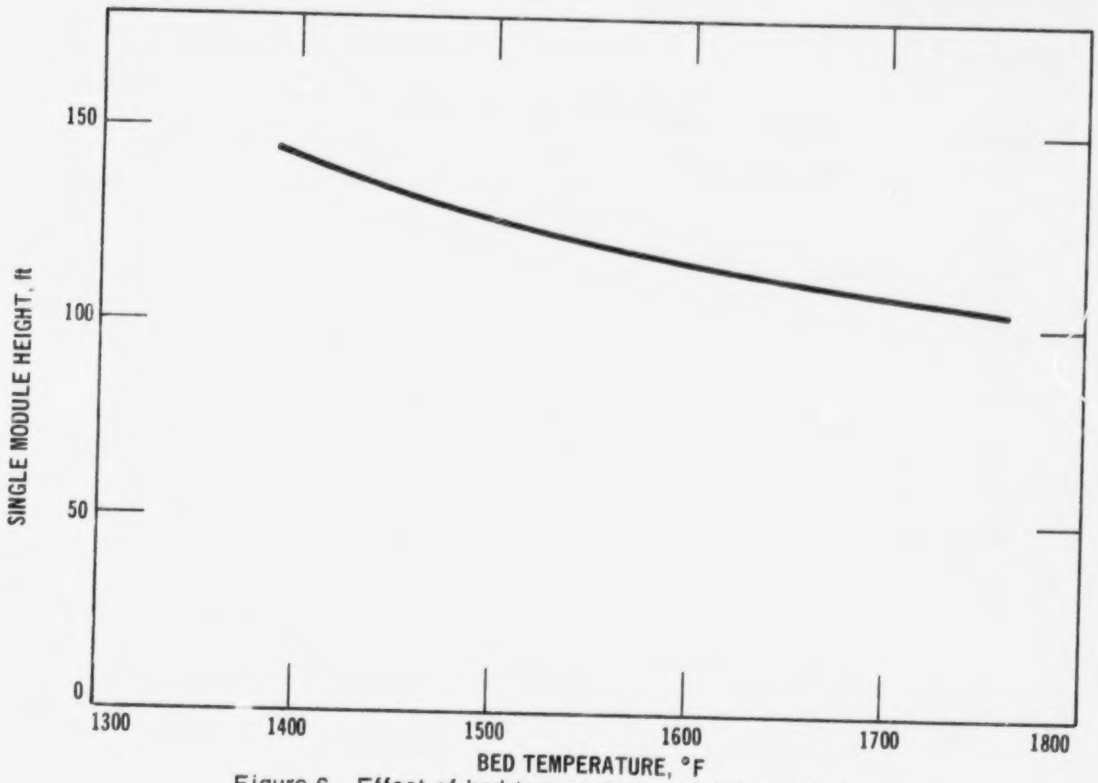


Figure 6. Effect of bed temperature on total module height.

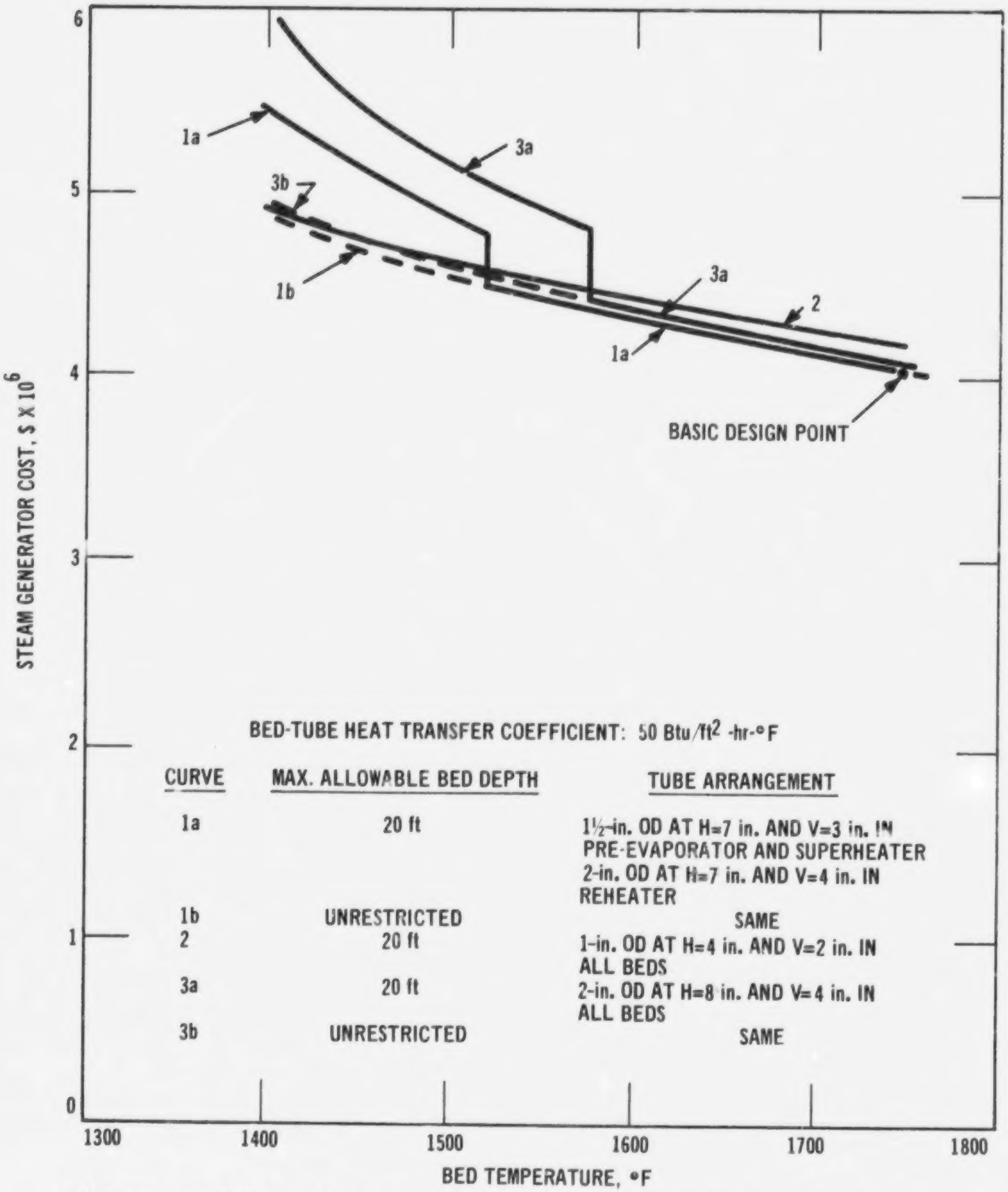


Figure 7. Effect of bed temperature on the steam generator cost of 318-MW plant (not including erection).

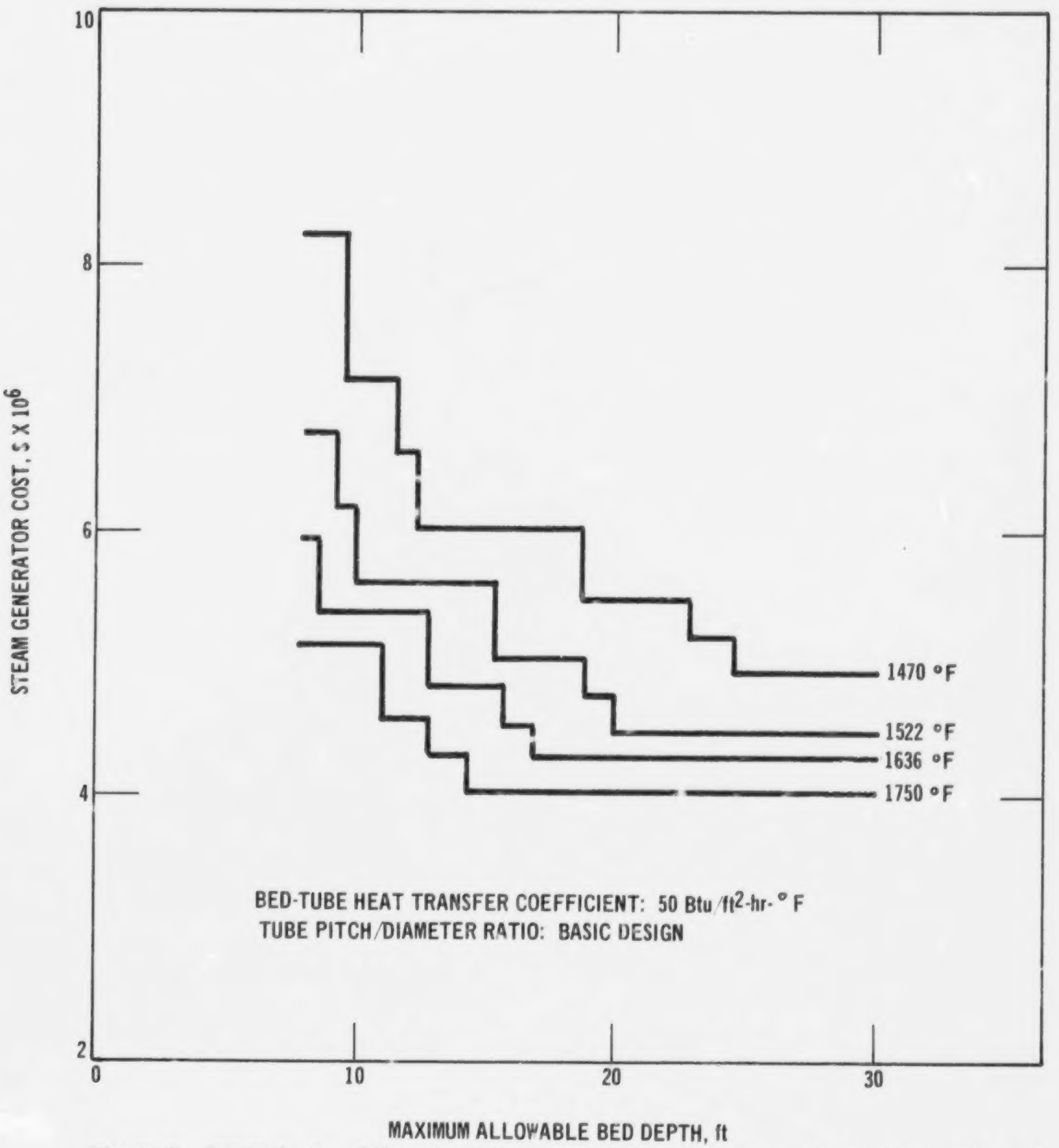


Figure 8. Dependence of the steam generator (318-MW) cost on the maximum allowable bed depth (not including erection).

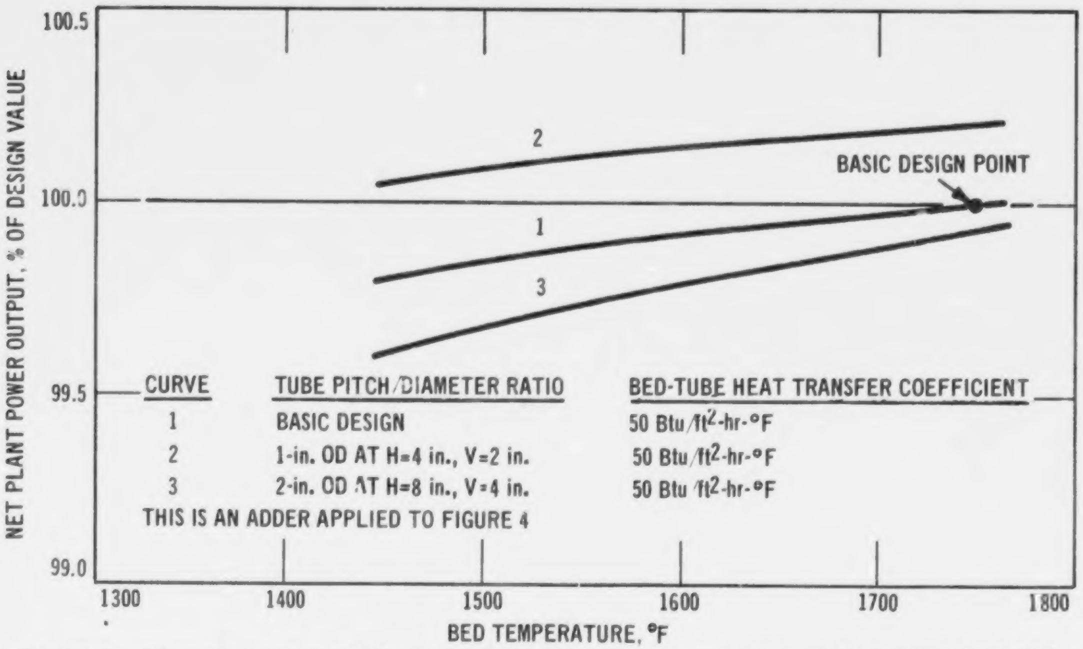


Figure 9. Change in net plant power output due to change in pressure drop across the bed.

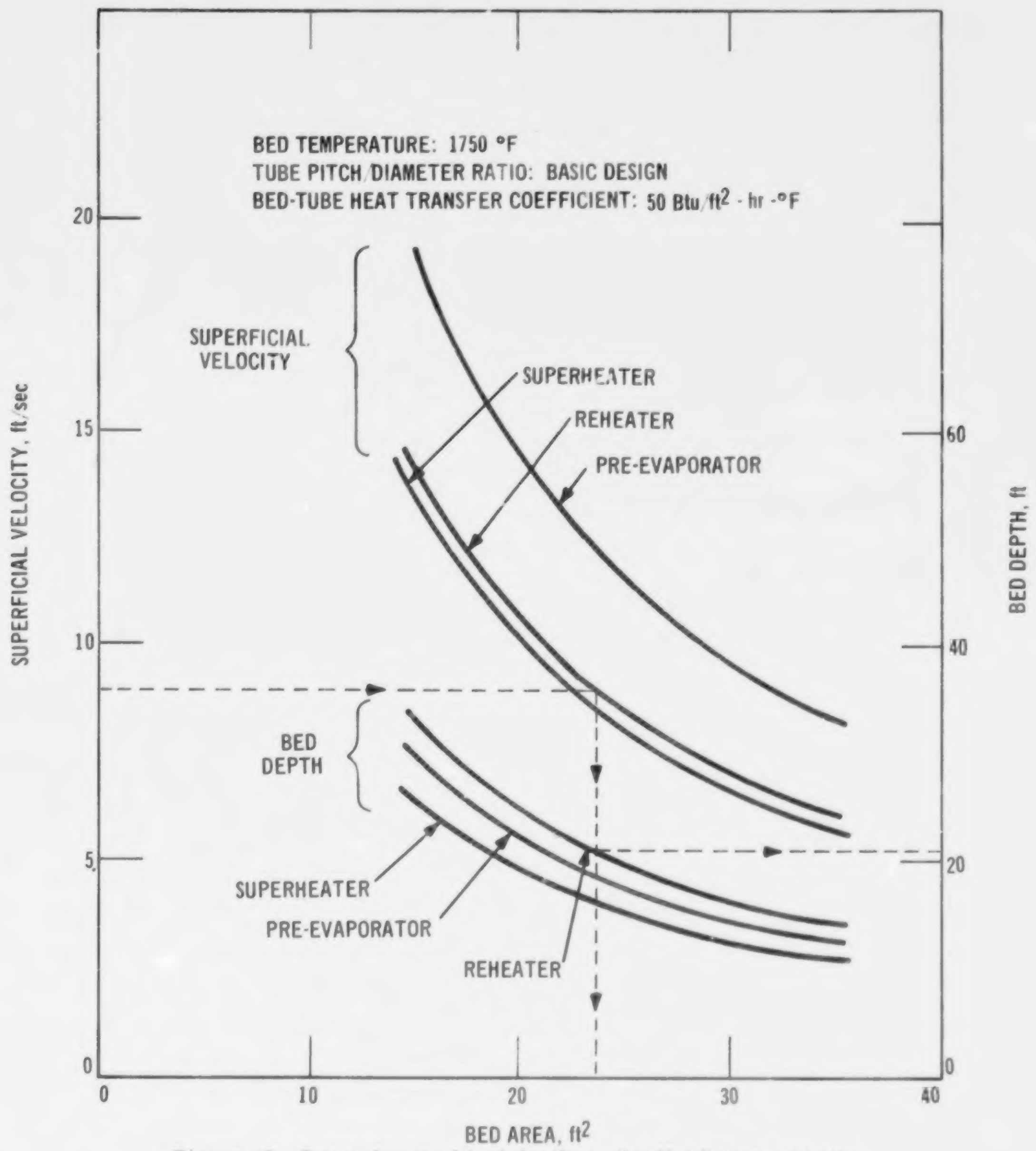


Figure 10. Dependence of bed depth on the fluidizing velocity.

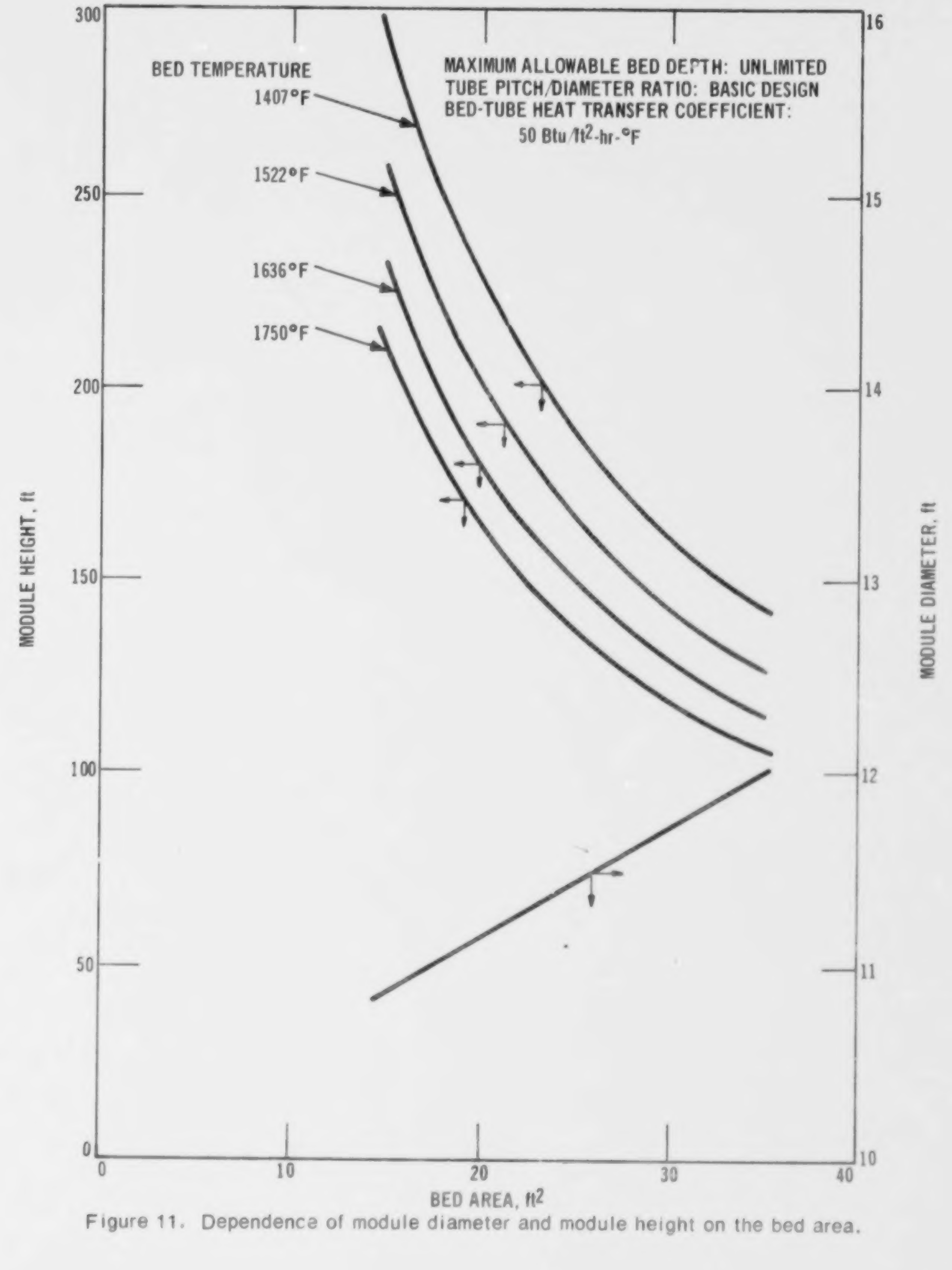


Figure 11. Dependence of module diameter and module height on the bed area.

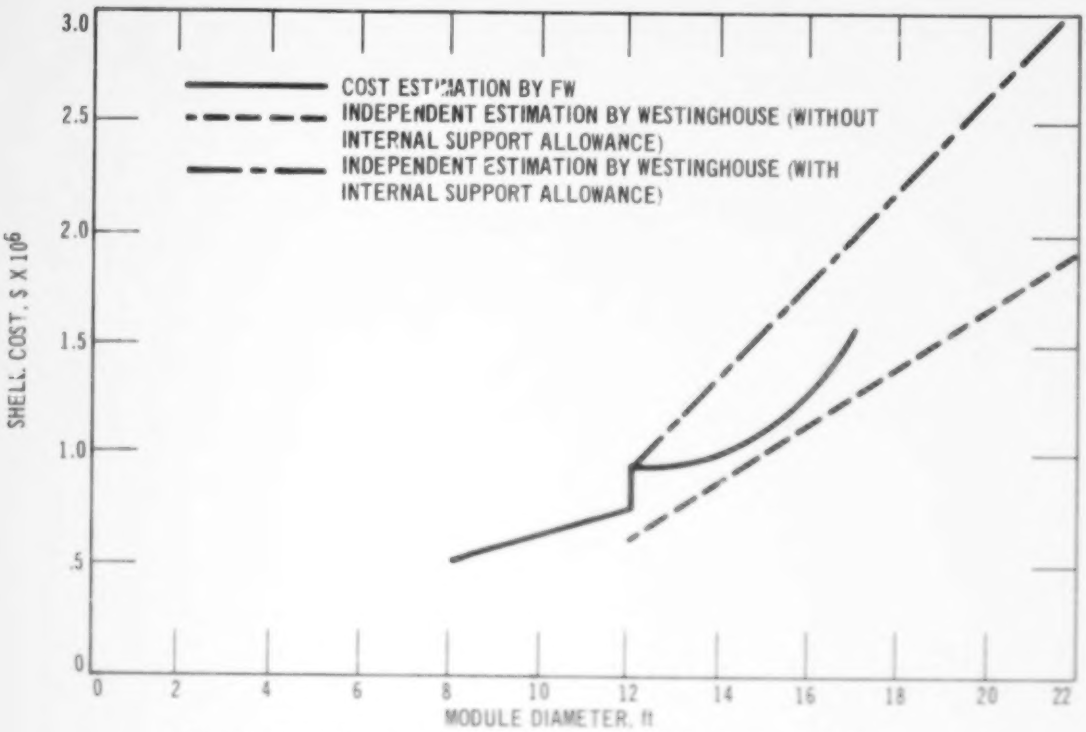


Figure 12. Shell cost (four modules) at basic design height.

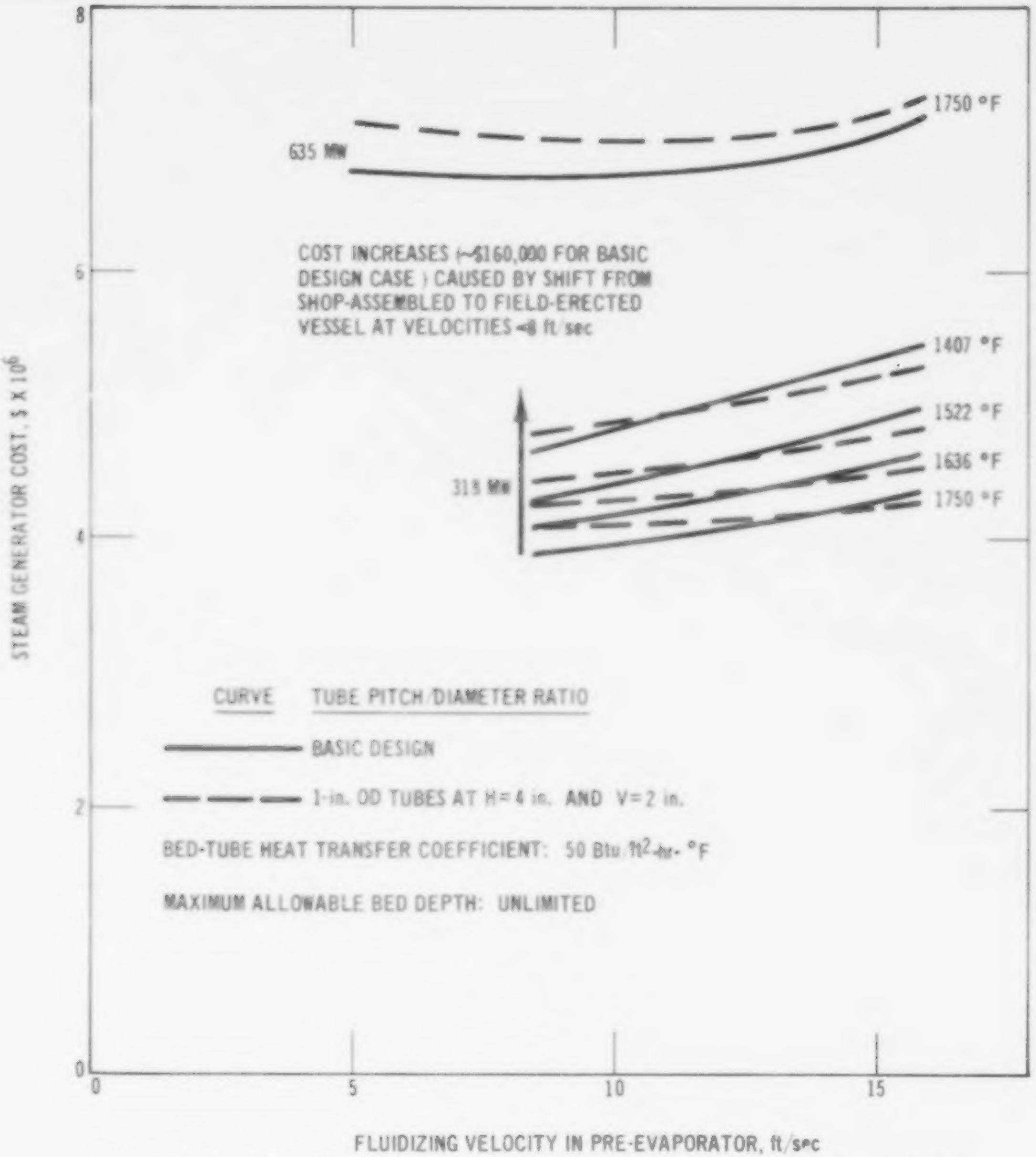


Figure 13. Dependence of the steam generator cost on the fluidizing velocity.

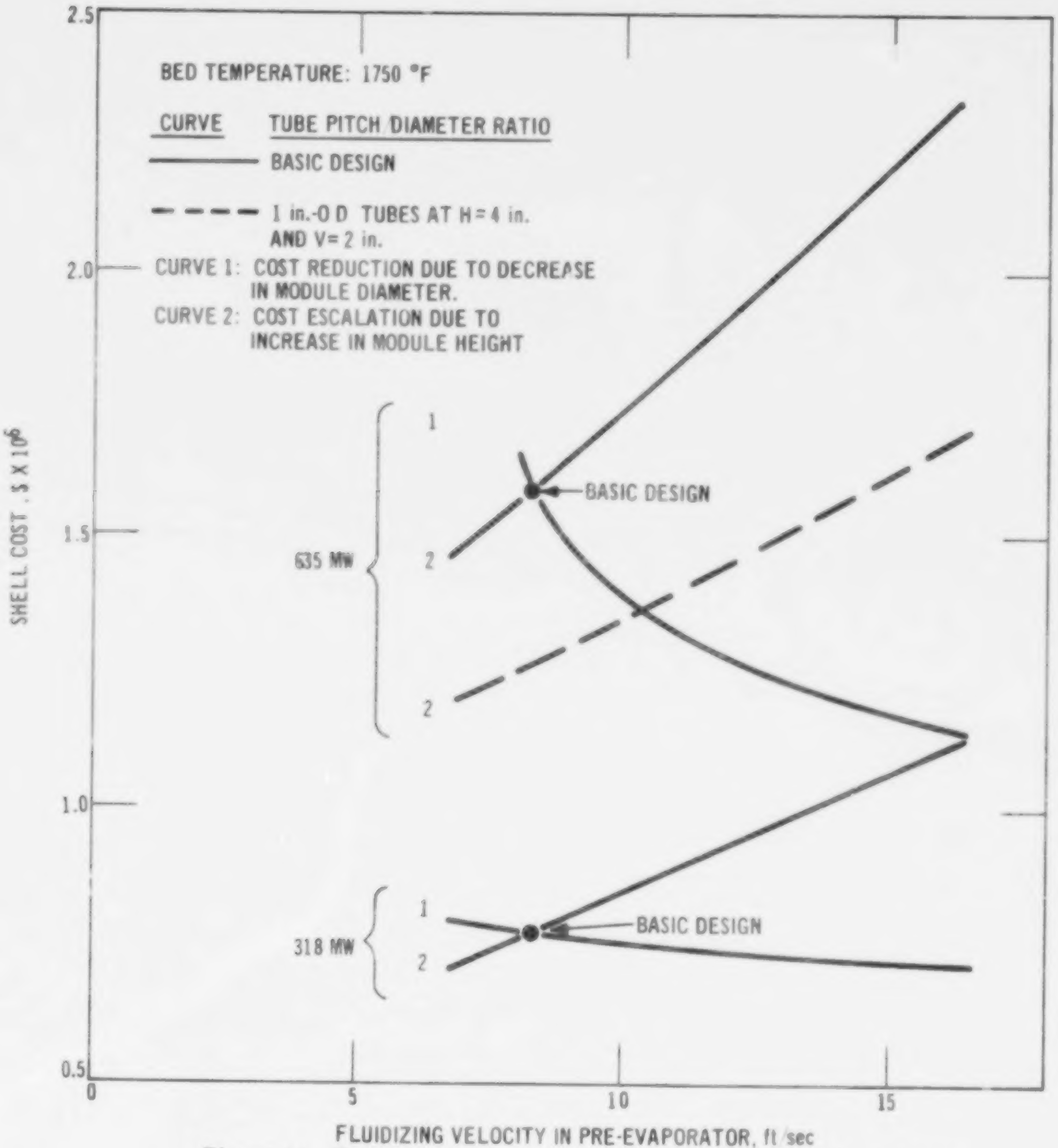


Figure 14. Dependence of the shell cost on the fluidizing velocity.

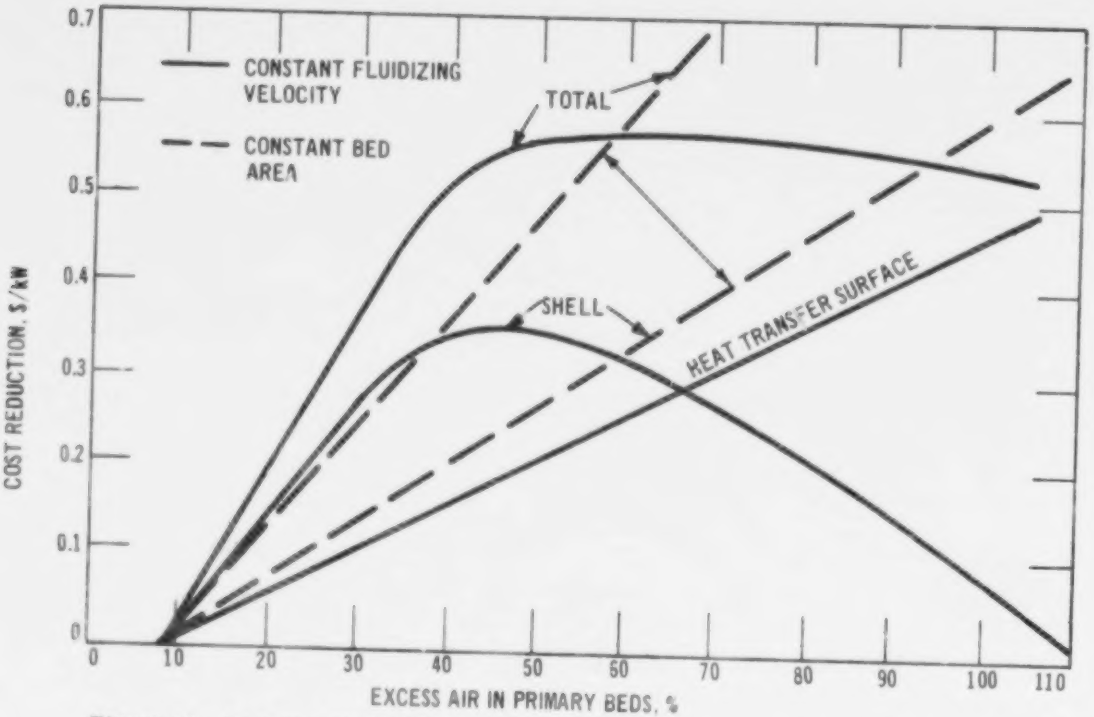


Figure 15. Cost reduction in heat transfer surface and pressure shell at different air flow rates.

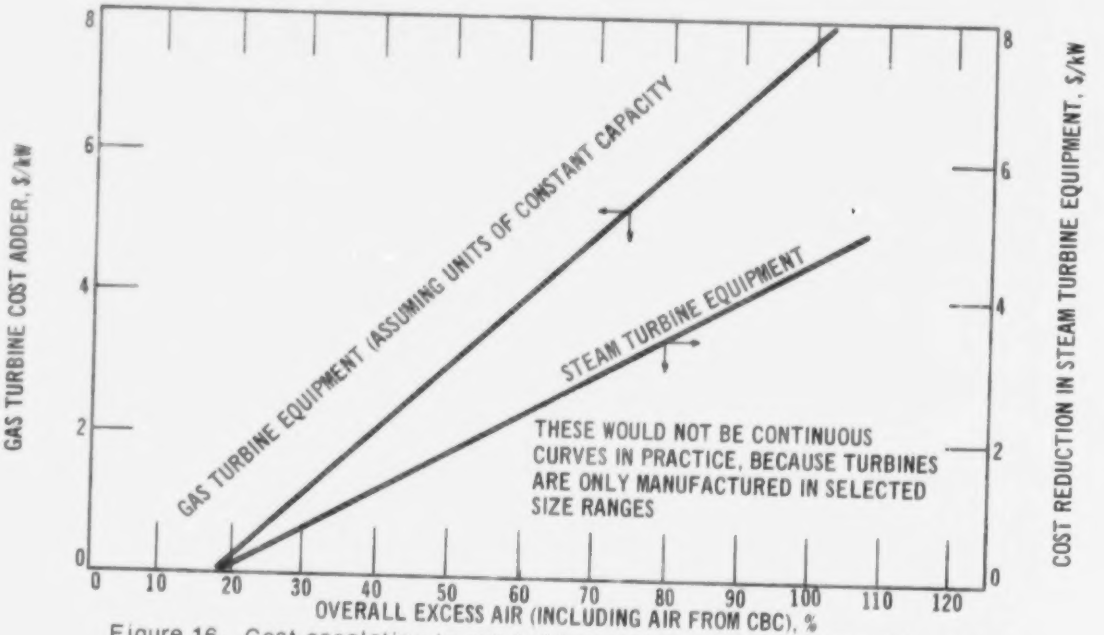


Figure 16. Cost escalation in gas turbine equipment and cost reduction in steam turbine equipment at different air flow rates.

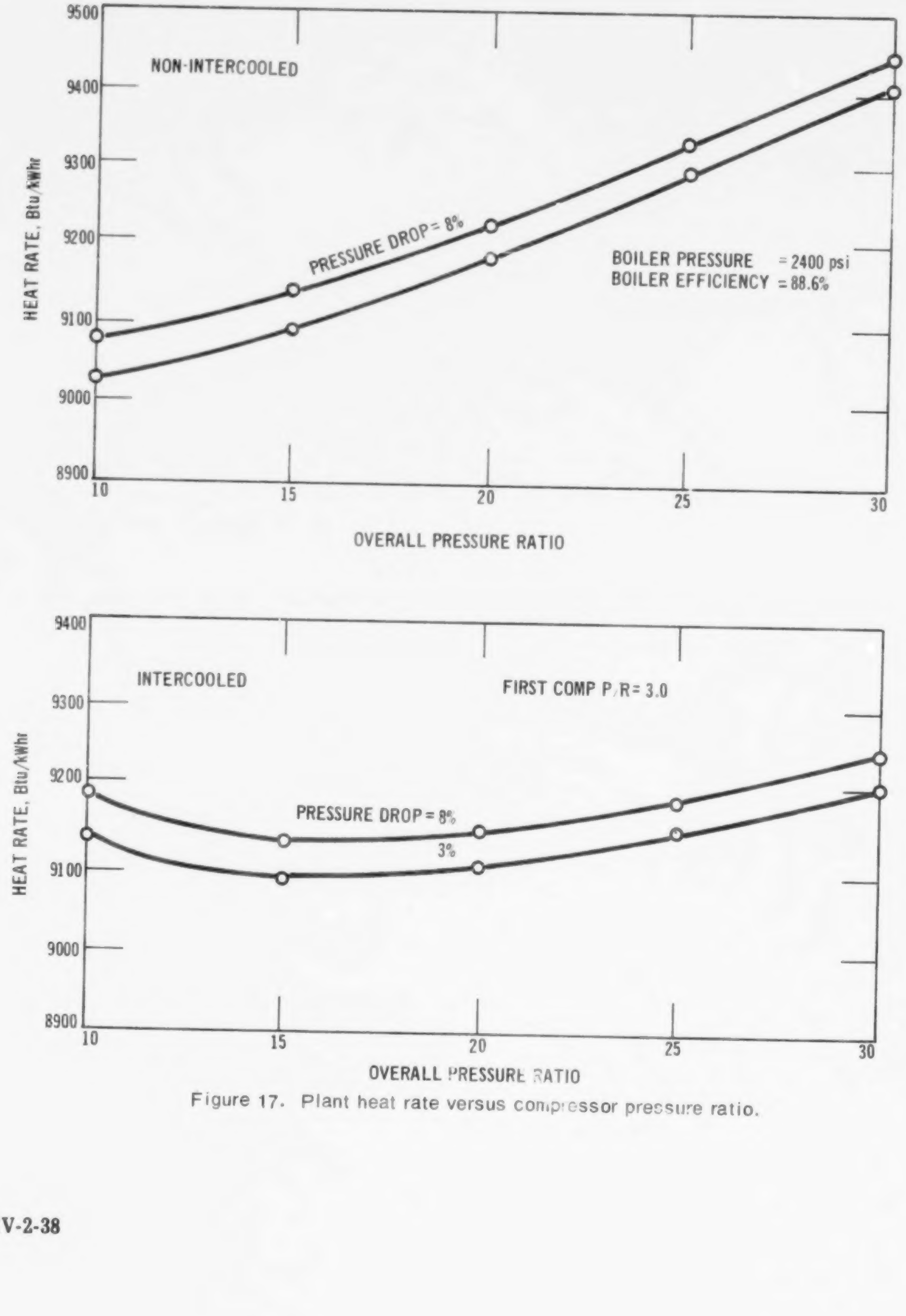


Figure 17. Plant heat rate versus compressor pressure ratio.

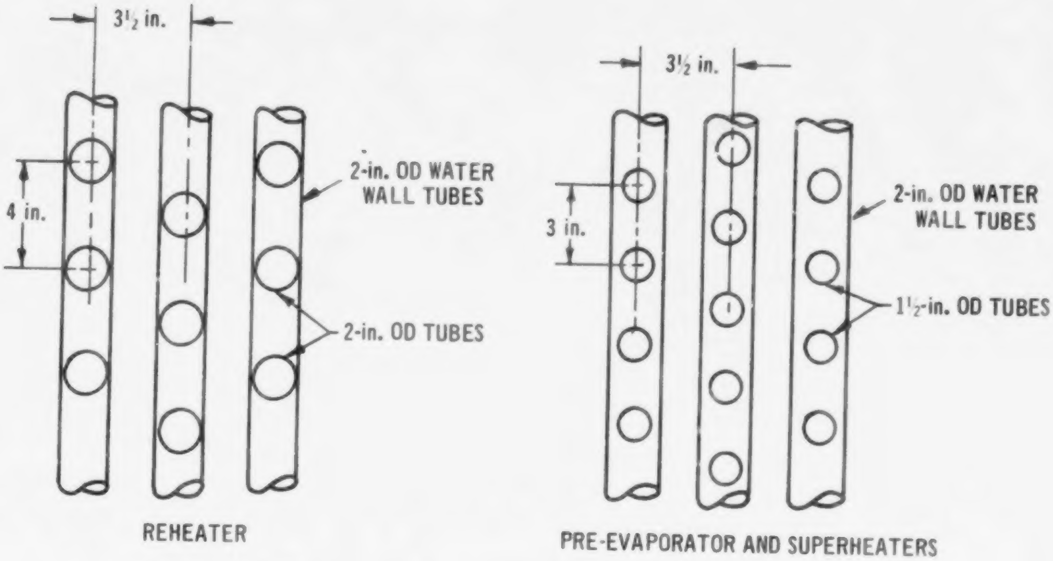

3½ in.
4 in.
2-in. OD WATER WALL TUBES
2-in. OD TUBES
REHEATER
3½ in.
3 in.
2-in. OD WATER WALL TUBES
1½-in. OD TUBES
PRE-EVAPORATOR AND SUPERHEATERS

Figure 18. Tube arrangement in basic design.

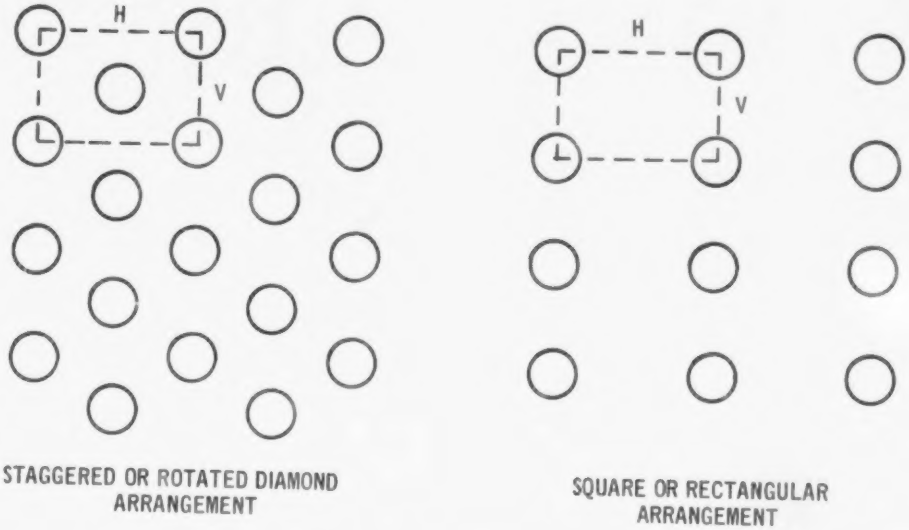

H
V
STAGGERED OR ROTATED DIAMOND ARRANGEMENT
H
V
SQUARE OR RECTANGULAR ARRANGEMENT

Figure 19. Definition for different tube arrangements.

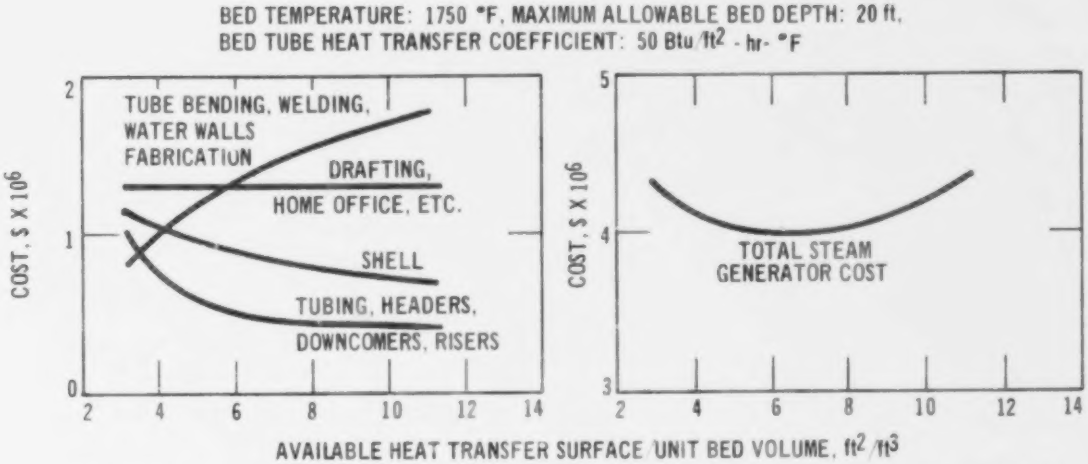


Figure 20. Change of the steam generator cost with heat transfer surface per unit bed volume (not including erection).

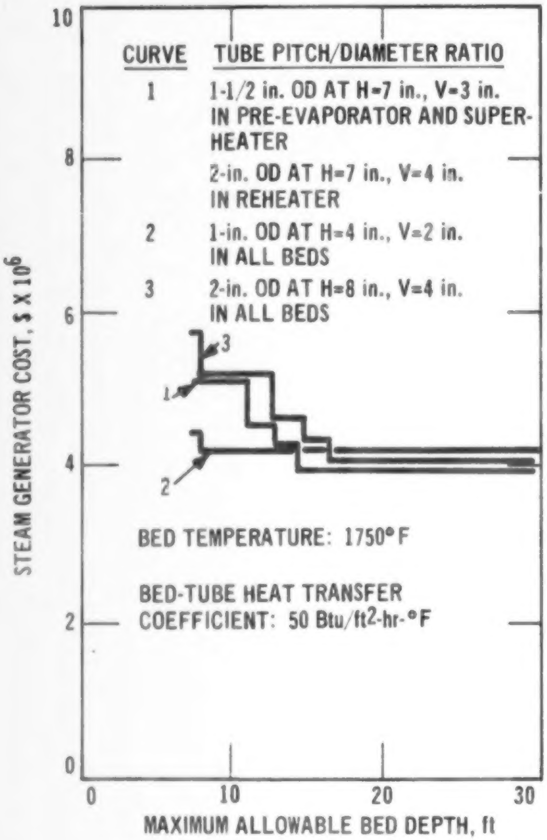


Figure 21. Dependence of the steam generator (318-MW) cost on the maximum allowable bed depth (not including erection).

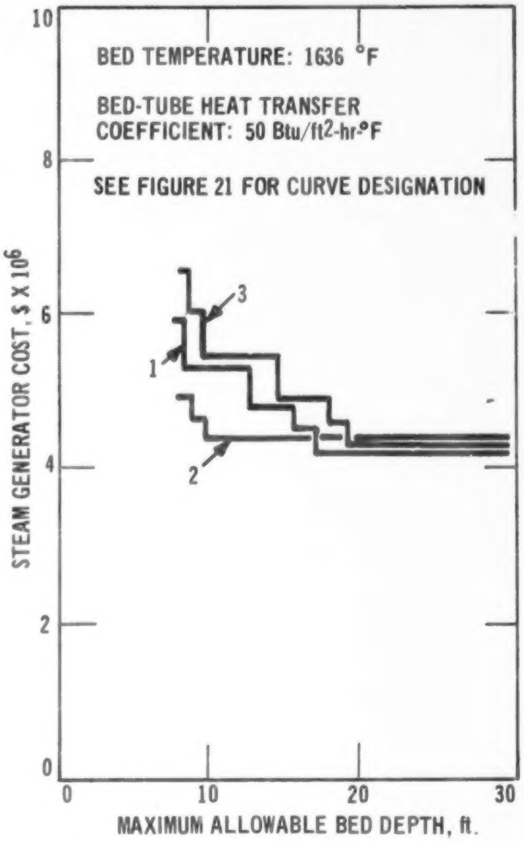


Figure 22. Dependence of the steam generator (318-MW) cost on the maximum allowable bed depth (not including erection).

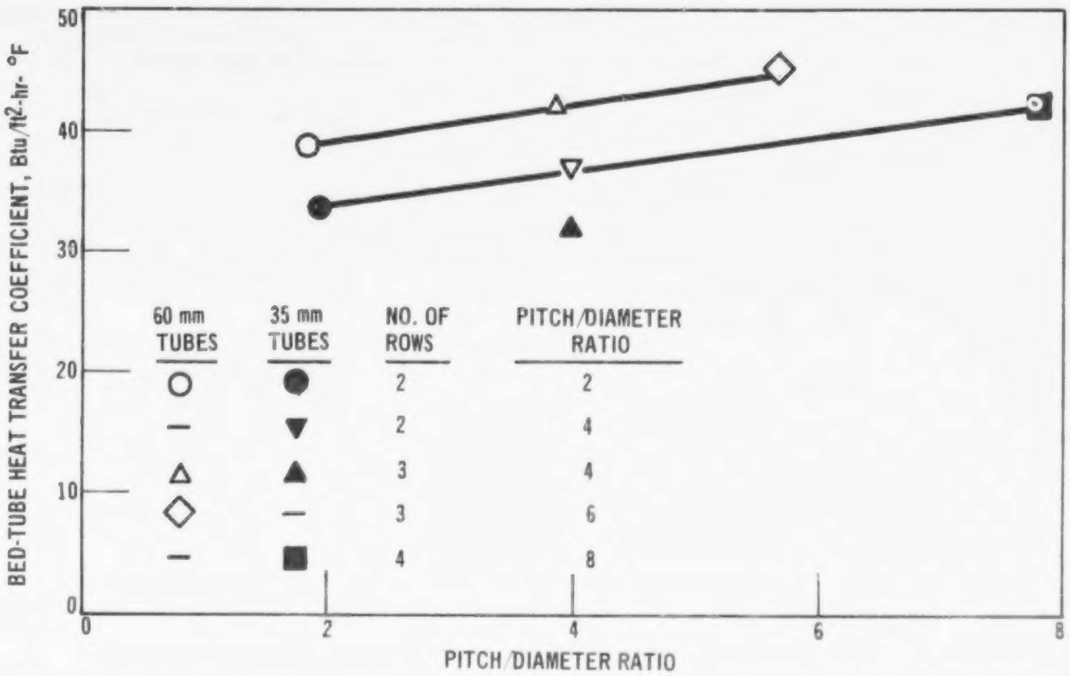


Figure 23. Change of bed-tube heat transfer coefficient with pitch/diameter ratio.[3]

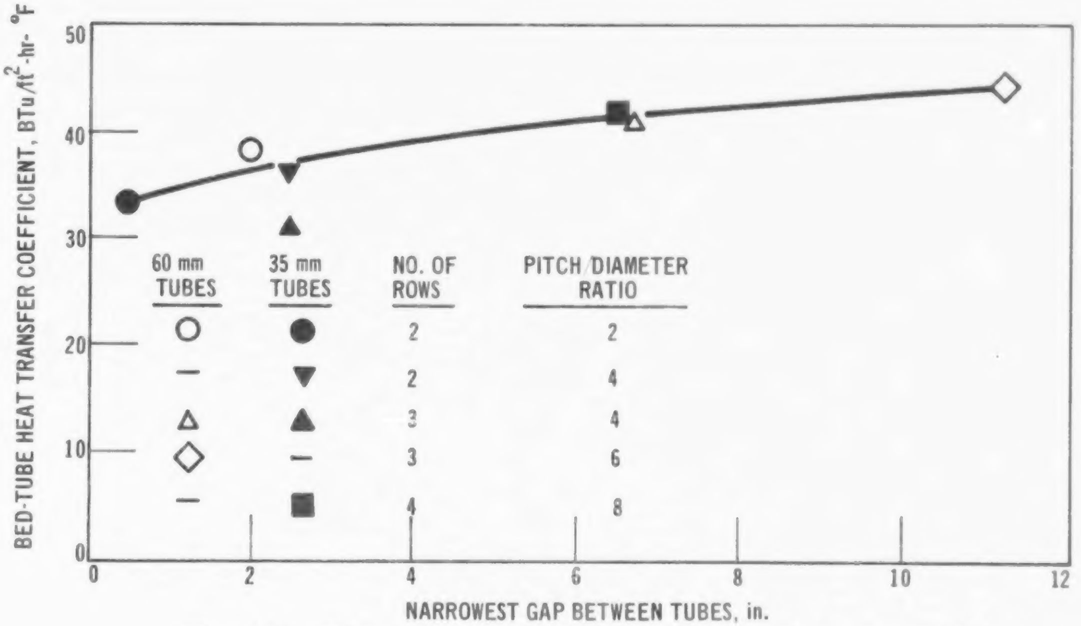


Figure 24. Change of heat transfer coefficient with tube spacing.[3,4]

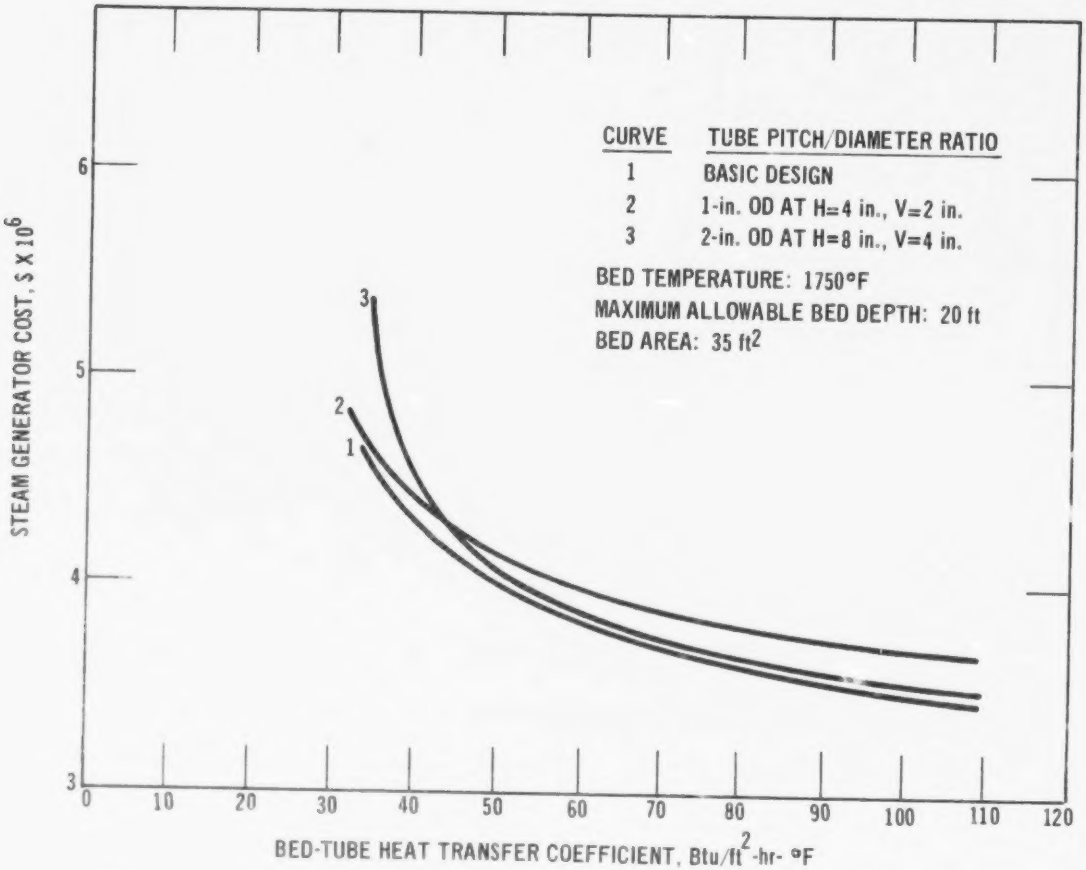


Figure 25. Effect of bed-tube heat transfer coefficient on the steam generator cost (not including erection).

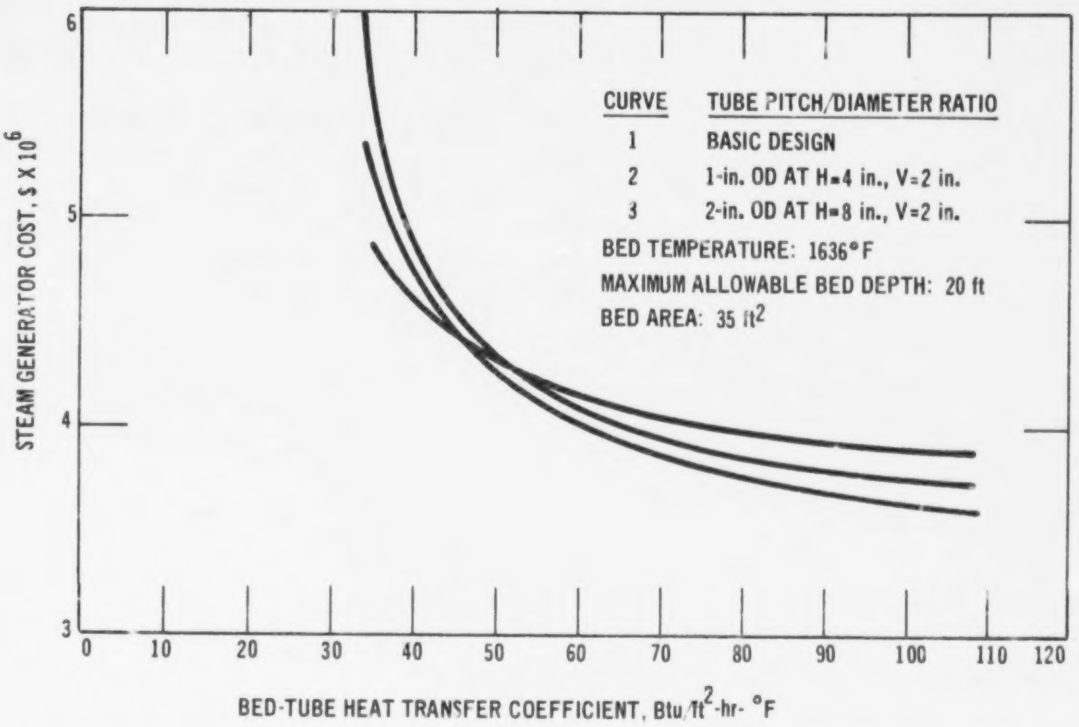


Figure 26. Effect of bed-tube heat transfer coefficient on the steam generator cost (not including erection).

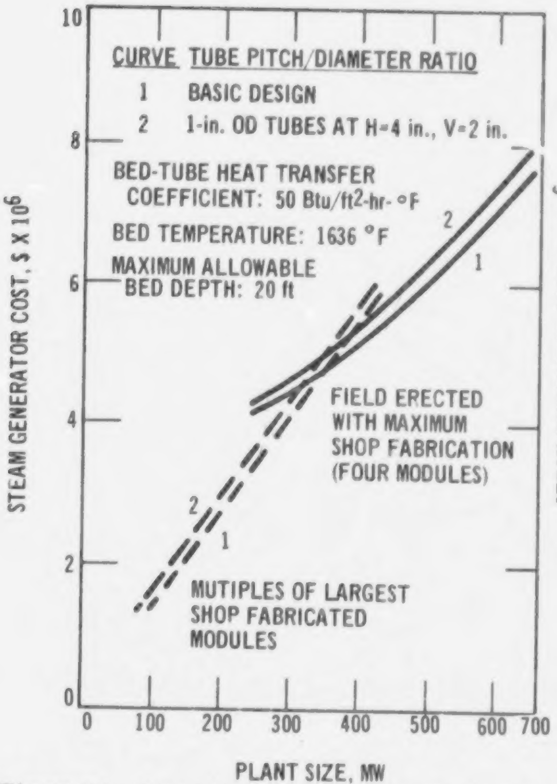


Figure 27. Dependence of the steam generator cost on the plant size.

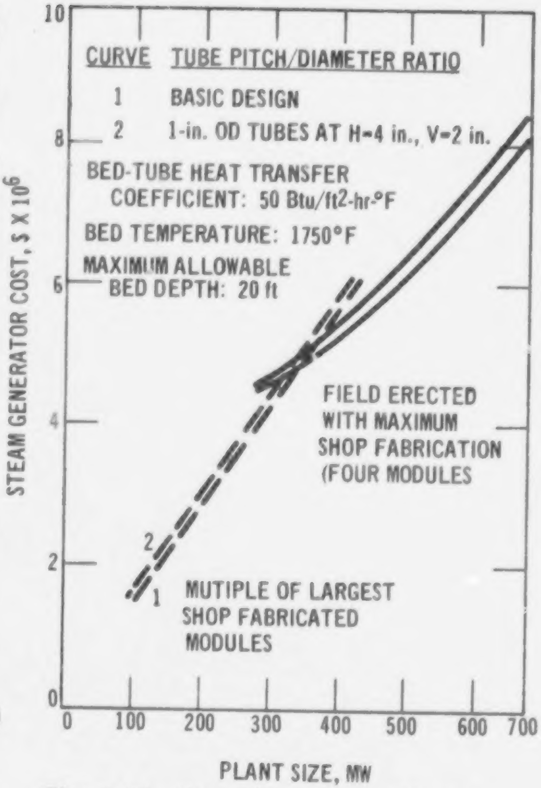


Figure 28. Dependence of the steam generator cost on the plant size.

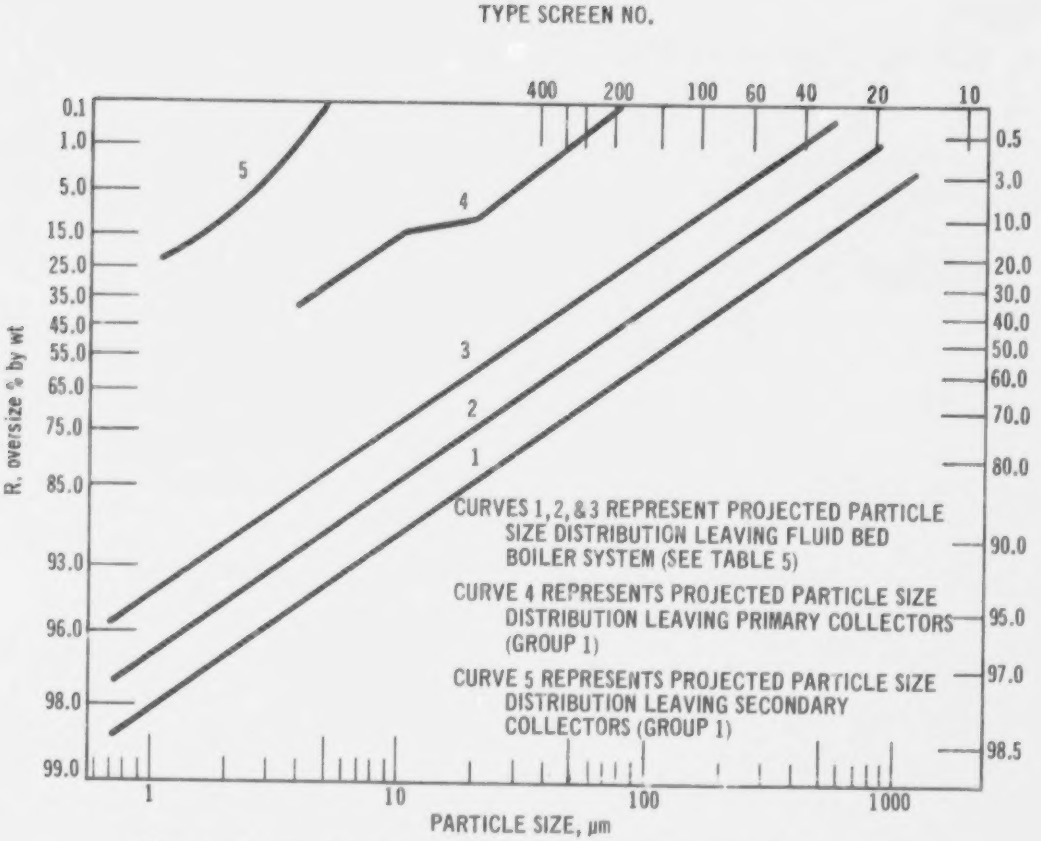


Figure 29. Particle size distribution for different gas streams.

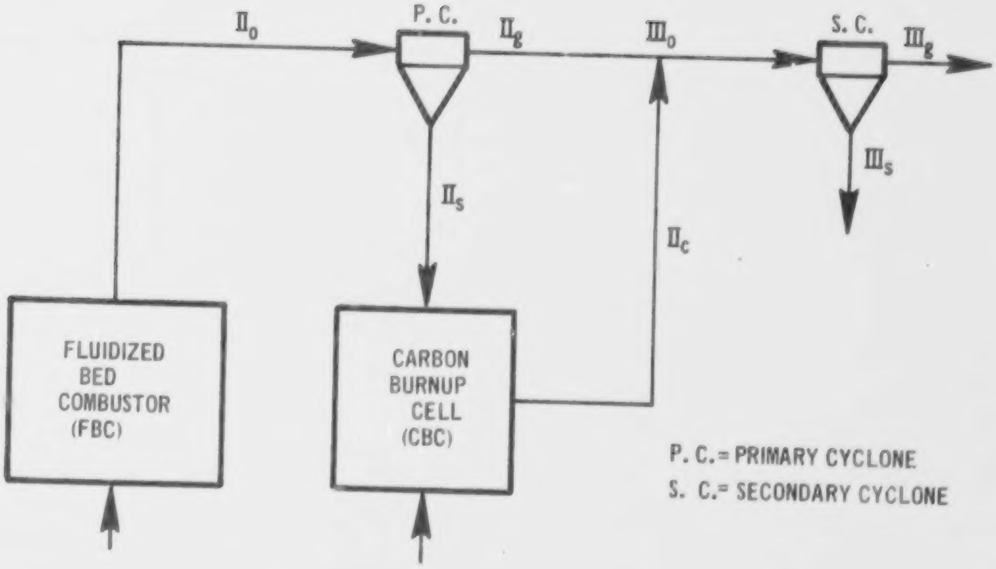


Figure 30. Flow diagram for particulate removal system.

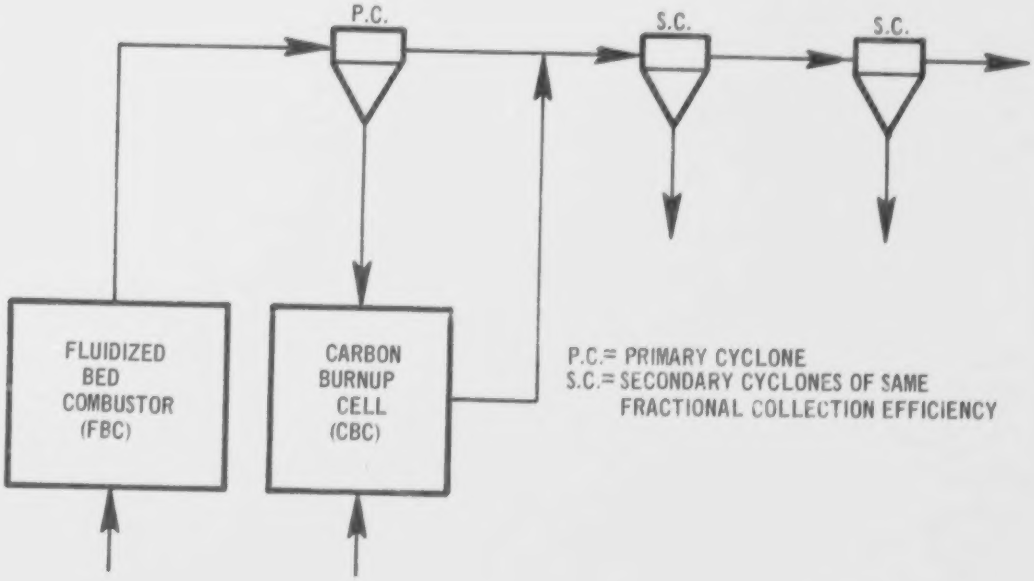

P.C.
S.C.
S.C.
FLUIDIZED BED COMBUSTOR (FBC)
CARBON BURNUP CELL (CBC)
P.C. = PRIMARY CYCLONE
S.C. = SECONDARY CYCLONES OF SAME FRACTIONAL COLLECTION EFFICIENCY

Figure 31. Flow chart for group 1, case 4.

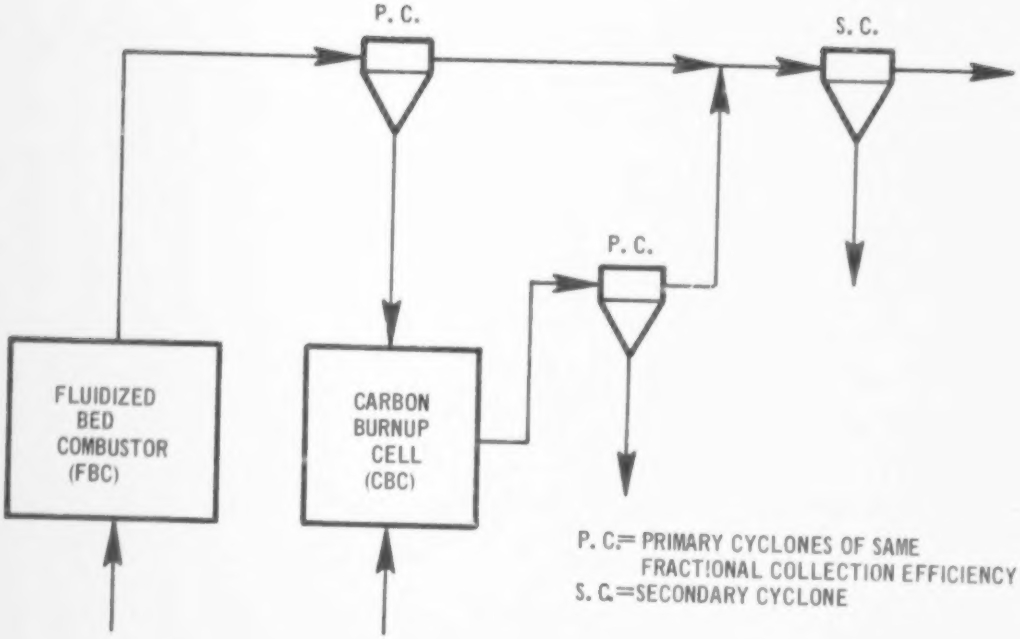

P. C.
S. C.
P. C.
FLUIDIZED BED COMBUSTOR (FBC)
CARBON BURNUP CELL (CBC)
P. C.= PRIMARY CYCLONES OF SAME FRACTIONAL COLLECTION EFFICIENCY
S. C.=SECONDARY CYCLONE

Figure 32. Flow chart for Group 1, Case 5.

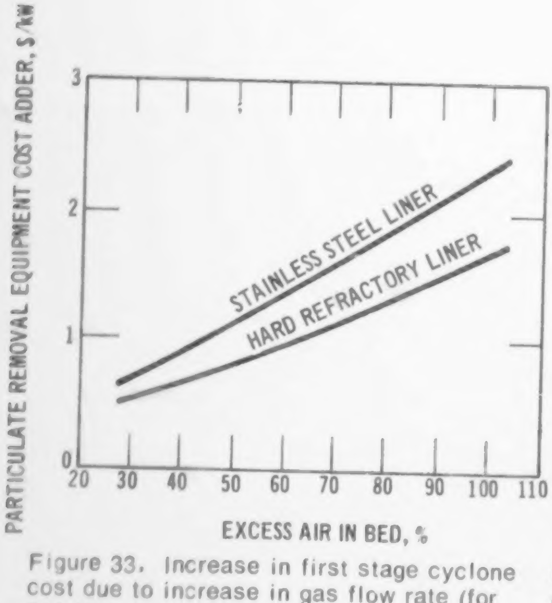


Figure 33. Increase in first stage cyclone cost due to increase in gas flow rate (for 318-MW plant).

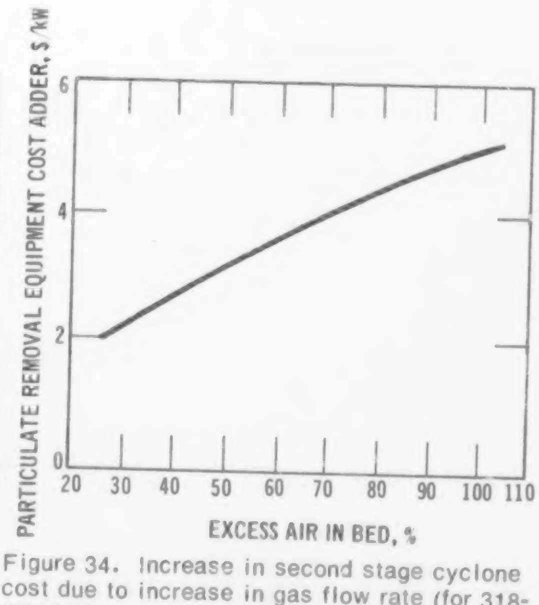


Figure 34. Increase in second stage cyclone cost due to increase in gas flow rate (for 318-MW plant).

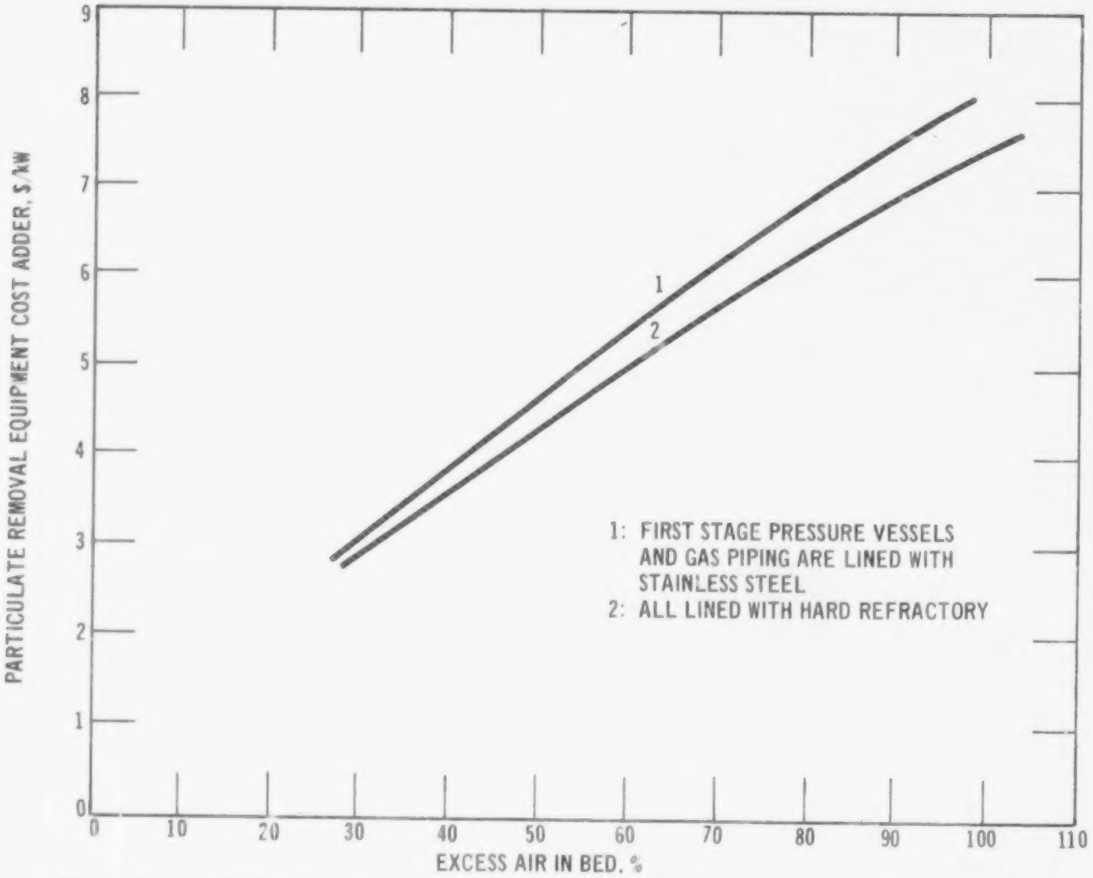


Figure 35. Cost increments for particulate removal system at different gas flow rates.

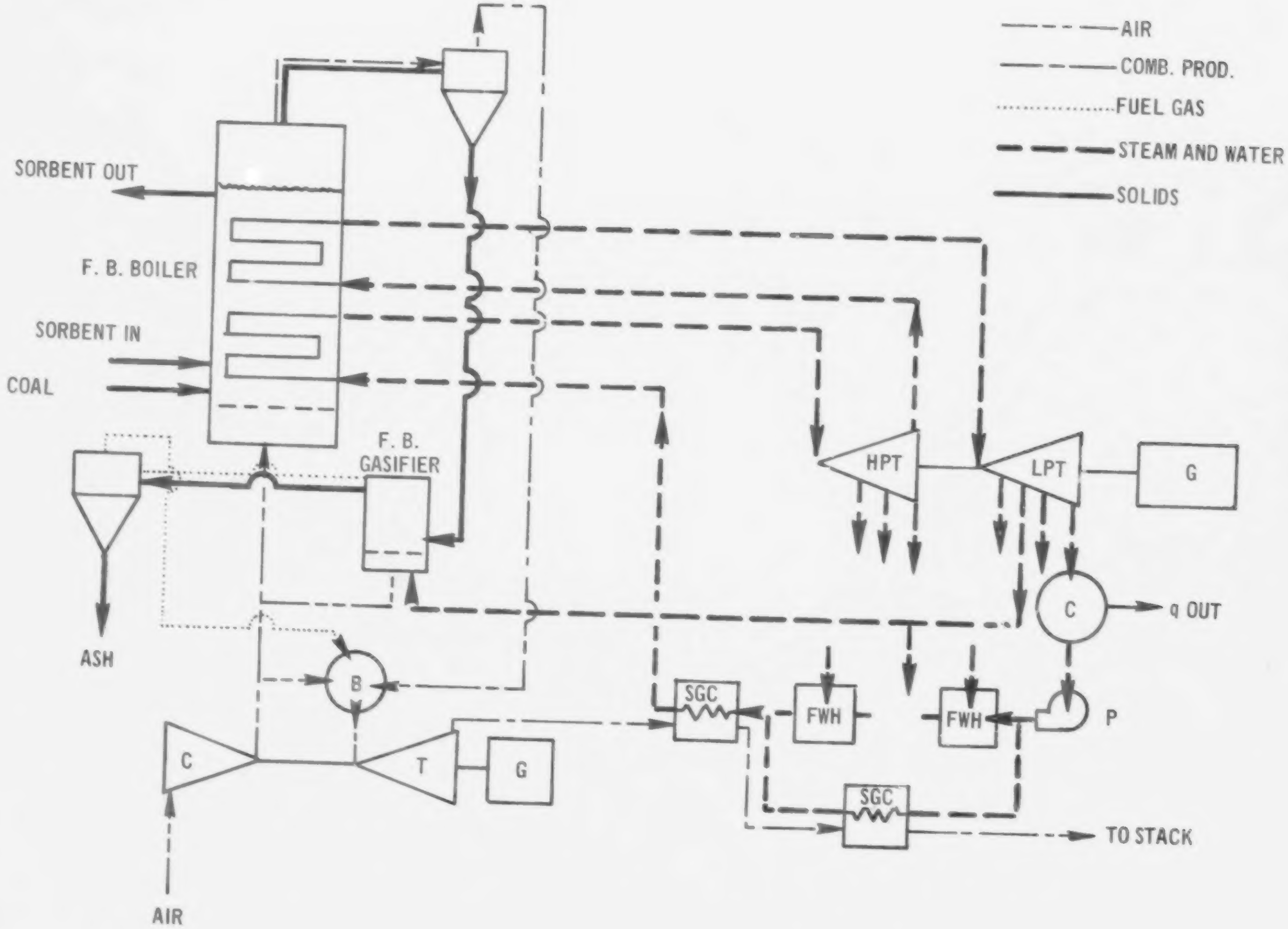


Figure 36. Pressurized fluid-bed combustion power plant with secondary combustor.

3. APPLICATION TO COMBINED CYCLE POWER PRODUCTION OF FLUID-BED TECHNOLOGY USED IN NUCLEAR FUEL REPROCESSING

B. R. DICKEY AND J. A. BUCKHAM

Allied Chemical Corp.

ABSTRACT

Fluid-bed processing is used extensively at the Idaho Chemical Processing Plant (ICPP) in the recovery of uranium from spent nuclear fuel elements. Fluid-bed denitration of uranyl nitrate solutions and fluid-bed solidification of radioactive waste solutions are used routinely in plant operations. In addition, current pilot-plant development of processes for recovering uranium from graphite-based fuels depends largely on fluidized-bed combustion.

During the course of fluidized-bed operations and development at ICPP, advantageous application of fluid-bed technology in areas other than nuclear fuel reprocessing has become apparent. Current and near-term fluidized-bed technology appears directly applicable to the conversion of the energy in wood wastes and municipal refuse to electric power. Fluidized-bed operations and process development at the ICPP and a concept for combined-cycle power production based largely on fluidized-bed combustion are discussed herein.

INTRODUCTION

Allied Chemical Corporation's Idaho Chemical Programs-Operations Office operates the Idaho Chemical Processing plant for the Atomic Energy Commission. The primary mission of the facility is to recover uranium from spent nuclear fuel elements in an economic and safe manner. Metallic clad nuclear fuels are dissolved in inorganic acids, and the uranium and fission product solutions are separated by solvent extraction. Uranium rich solutions are denitrated and solidified to uranium oxide; fission product solutions are calcined to a mixture of metallic and fission product oxides.

Fluidized-bed processing is used in the solidification of fission product solutions and in denitration of uranium rich solutions. Fluid-bed solidification of radioactive waste solutions has been used since December 1963. Denitration by thermal decomposition in a fluidized bed was started in 1971. Both waste calcination and denitration are endothermic processes. In-bed combustion of kerosene supplies the heat for waste calcination; heat for fluid-bed denitration is obtained from wall-mounted electrical heaters.

The capacity to recover enriched uranium from graphite-based fuels will be required in the near future. Pilot-plant development of processes for separation and recovery of uranium from these fuels is well underway. The heart of these processes is the fluidized-bed combustion of the graphite matrix.

In the course of plant operation and process development using fluid-bed technology at the ICPP, concepts based on the application of fluid-bed technology in non-nuclear areas have evolved. A proposed application of current interest is the recycle of wood wastes and municipal refuse through the conversion to electrical power. The proposed process is based on a combined gas turbine-steam turbine cycle; the key to producing power at high thermal efficiencies in the proposed concept is a fluidized-bed combustor.

OPERATIONAL FLUID-BED PROCESSES AT ICPP

Waste Calcining Facility

Radioactive wastes resulting from nuclear fuel reprocessing can be solidified by various methods that have been under investigation over the past 20 years; however, only one process has been demonstrated on a production basis. The Waste Calcining Facility (WCF) at the Idaho Chemical Processing Plant is the first production facility in the world for converting aqueous radioactive wastes to solids using the fluidized-bed calcination process.

A schematic flow sheet for the WCF is shown in Figure 1. The heart of the process is the 4-ft diameter calciner vessel where the radioactive aqueous wastes are continuously atomized into the fluidized bed of oxide particles by an external-mix pneumatic atomizing nozzle. In the fluidized bed, which is maintained at 500°C, water evaporates, the metallic salts are converted to their corresponding oxides or fluorides which are deposited layerwise on the spherical bed particles. The solid particles are withdrawn continuously from the calciner vessel to maintain a constant bed height and are transported pneumatically to stainless steel solid storage bins adjacent to the calcining facility.

The off-gas leaving the calciner vessel passes through a dry cyclone where the majority of elutriated fines are removed and pneumatically transported to the solids storage bins. The off-gas then passes through a wet scrubbing system consisting of a quench tank, venturi scrubber, cyclone separator, and a demister. Here, the off-gas is contacted with a nitric acid scrubbing solution which removes the majority of the remaining solids. The scrubbing solution is recirculated continuously, and any accumulation is recycled to the waste feed tank.

Heat for the endothermic calcination reactions is supplied by in-bed combustion. In-bed combustion consists of atomizing a hydrocarbon fuel (kerosene) with pure oxygen directly in the fluidized bed. Startup of the process is achieved by heating the fluidized bed to temperatures in the range of 360 to 400°C using preheated fluidizing air. A nitrate-containing waste is then injected through a separate waste atomizing nozzle, followed immediately by the injection of the fuel-oxygen mixture through the fuel atomizing nozzle. Ignition of the fuel-oxygen mixture is spontaneous at temperatures above 335°C in the presence of nitrates. After the startup, the wastes are calcined at temperatures in the range of 400 to 500°C during routine operation. The advantage of this method of heating is that no heat transfer surfaces are involved that can foul or limit capacity; the heat flow paths of in-bed combustion heating and an in-bed heat exchanger are compared in Figure 2.

Denitration facility

The denitration process is based on the thermal decomposition of uranyl nitrate solution to uranium trioxide. In fluidized-bed denitration, solution is continuously sprayed through an air atomizing nozzle into heated fluidized bed of UO_3. The bed temperature is 300°C; the pressure immediately above the support plate is approximately atmospheric. Granular product is continually withdrawn into a product collection vessel. The process off-gas--consisting mainly of fluidizing air, water vapor, and oxides of nitrogen--flows through a filtering section consisting of three sintered metal filters which remove over 99.9 percent of the entrained and elutriated UO_3 dust particles. The filters are blown back intermittently, and the fine particles serve as seed particles for particle growth. A schematic flow-sheet of the fluid-bed denitration process is shown in Figure 3.

FLUID-BED BURNING PROCESSES

Pilot-plant development of a fluidized-bed combustion process for separating uranium from spent graphite-matrix nuclear fuels has been in progress since January 1966 at ICPP.[2] This unique combustion process is required because graphite, unlike the metallic cladding of the more conventional nuclear fuels, is not readily dissolved in common inorganic acids. As the graphite matrix is removed by combustion, the uranium and other metals are converted to their oxides. Dissolution of the resulting uranium oxide, U_3O_8, completes the burn-leach head-end process. The uranium is separated from fission products and other impurities by conventional solvent extraction.

The fuel consists of uranium dicarbide microspheres coated with pyrolytic carbon dispersed in a graphite matrix. A protective coating of niobium carbide is present on some of the surfaces of the fuel elements. Although aluminum or stainless steel orifices are present in some elements, there are only three constituents of consequence to the combustion process: uranium, niobium, and carbon.

The fluidized-bed combustion process developed at ICPP for nuclear rocket fuels involves the following concepts: (1) charging of whole fuel elements to a fluidized bed of inert alumina particles, (2) combustion of essentially all carbon, (3) oxidation of uranium and niobium carbides, and (4) elutriation of uranium and niobium oxides from the burner.

Requirements for the fluidized-bed burning and elutriation processes are: (1) combustion of at least 95 percent of the matrix graphite and pyrolytic carbon, (2) conversion of the uranium dicarbide microspheres and niobium carbide to elutriable particles by oxidation and attrition, (3) negligible attrition and elutriation of the inert bed material (α-alumina), and (4) adequate heat dissipation for control of bed temperatures. If "steady-state" conditions can be achieved, all of the uranium and niobium charged to the burner will be elutriated in the burner off-gas. The amount of alumina and unburned carbon carried overhead with the U_3O_8 product must be minimized.

The fluidized-bed burner segment of the Graphite Fuels Pilot Plant (GFPP) (Figure 4) was constructed to permit studies of the combustion process. The burner and leaching equipment, located on two adjacent modules, can be operated independently or simultaneously.

Major components of the burner module are a fluidized bed for burning the fuel and a dry product collection system for filtering and retaining particulates from the burner off-gas. The dry product collection system is bypassed when direct introduction of the burner product into the leaching equipment is desired.

Several burner-designs were used in the course of pilot-plant development. The final burner design, the concentric fluidized-bed burner, is shown schematically in Figure 5. A 4-in. diameter, 4-1/2-ft long first-stage burner is located concentrically inside a 6-in. diameter vessel which extends 8-1/2 feet above the top of the first stage. Fluidized beds are contained in the 4-in. diameter first stage, the surrounding annular space, and in the 6-in. diameter section above the first stage. The wall of the upper 2 feet of the first-stage burner is slotted to allow particle mixing between the annular and inner beds.

The concentric fluidized-bed design was proposed on the basis of potential increased heat transfer rates from the wall of the inner first-stage burner. The outer wall of the first-stage burner in an earlier two-stage fluidized-bed burner was cooled by forced-air convection; this proved to be adequate under normal conditions but inadequate in the event of a temperature excursion. Substitution of the annular bed for the forced convection system was expected to increase the heat transfer coefficients at the first-stage wall by an order of magnitude. The annular fluidized bed provides the added advantage of secondary containment should melt-through of the inner vessel occur.

Tests conducted with a 6-in. diameter glass column containing the actual 4-in. diameter inner bed further showed that increased heat transfer could also be expected from particle mixing between the inner and annular beds. The intensity of slugging in the inner bed was greatly reduced in the upper 2 feet due to the transfer of gas and particles between the inner and annular beds.

The anticipated improvement in heat transfer using the concentric fluidized-bed burner has been realized. The concentric-bed design has proved superior to an original two-stage burner with respect to heat transfer and temperature control. No temperature excursions have occurred in any of the experiments. Temperature differentials within the first stage have ranged from a normal spread of 25 to 45°F to a maximum spread of 75°F.

Combustion efficiencies equal to or greater than the required 95 weight percent have been obtained in the two-stage concentric fluidized bed over the following ranges of operating variables:

1. Nominal bed temperature—1400 to 1500°F.
2. Fluidizing-gas composition—80 to 100 percent oxygen to both stages.
3. Mean fluidizing velocities (average of inner and annular fluidizing velocities) — 1.05 to 1.50 ft/sec.
4. Fuel - charging rates--up to 33 kg graphite/hr-ft^2.

APPLICATION OF FLUID-BED COMBUSTION TO COMBINED-CYCLE POWER PRODUCTION FROM WOOD WASTES AND MUNICIPAL REFUSE

Problem Definition

At present, approximately 250 x 10^6 tons (190 x 10^6 tons of which are collected) of residential, commercial, and institutional wastes are produced in the United States each year. The per capita generation of such wastes is rapidly increasing; combined with an expanding population, the magnitude of the problem may double by the turn of the century. The disposal of refuse is becoming a critical problem for communities, especially those in the heavily populated parts of the country.

The lumber industry, particularly the small-to-medium size mills, has a similar waste disposal problem. Approximately 5 x 10^6 tons of wood wastes are produced in the United States per year. The volume of waste makes landfill disposal impractical; the wastes are usually burned in inefficient teepee burners. While economical, such systems cause localized air pollution by emitting smoke, particulate matter, and partially-oxidized chemicals. Failure to meet the stringent air pollution standards (now being introduced) will prevent future operation of teepee burners.

Recycle of Solid Waste By Energy Production

Through regional planning and cooperation, wood waste materials and municipal refuse would be transported to a central location and burned in a highly efficient pressurized fluid-bed burner. Heat released in the bed would be used to generate steam within an in-bed heat exchanger; both the steam and high temperature off-gases (1400 to 1500°F) would generate electrical power using steam and gas turbine cycles, respectively. Removal of heat by generating steam in an in-bed heat exchanger would also minimize the amount of excess air normally required to control the bed temperature; thus, the volume of off-gas requiring cleanup would be reduced. Technology required for plant-scale process demonstration is either already established or in the latter stages of development.

Advantages of the proposed system are:

1. Significant reduction in present air and solid waste pollution (to meet present and future standards).
2. Decreased land requirements for disposal.
3. Conservation of natural resources (fossil and nuclear fuels) by recycling wastes to produce electrical power.
4. Decreased cost of waste disposal.

While the concept is one in which both wood and municipal wastes are available, successful development and plant-scale demonstration of the proposed concept could lead to use in areas where either type of fuel predominates. The concept should find wide application throughout the Pacific Northwest, the North, and the Southeast.

A schematic flowsheet of the conceptual plant is shown in Figure 6. Although the plant would be designed to process wood waste and municipal refuse as the principal fuels, the basic concept is compatible with other waste (e.g., industrial wastes, sewage sludge, etc.) as fuel. In the future, the plant could be modified to accept feed in the form of low sulfur coal as fuel in the event that advancing technology results in better use of the waste materials.

After the required preparation (e.g., screening, shredding, and storage), the waste is partially dried and charged to a fluid-bed burner operating at 150 psia and in the temperature range of 1400 to 1500°F. Combustion occurs in an inert bed of sand; the efficient solids and gas contact in the bed results in rapid and complete combustion. Ash and noncombustibles are continually withdrawn from the burner; some particulate is carried overhead in the off-gas.

Steam is generated from condensate passing through the tubes of an in-bed heat exchanger. The superheated high pressure steam then flows to a steam turbine-generator to produce electrical power. Flue gases from the fluid bed are cleaned of particulate matter using a combination of cyclones and high efficiency filters (ceramic or sintered metal). The clean flue gas then flows to a gas turbine to generate additional electric power.

The concept of combining gas and steam turbine cycles in a system for generation of electric power is not new, and the high thermal efficiencies possible in such cycles are being demonstrated in utility power stations. The San Angelo Station of West Texas Utilities, for example, has achieved 41 percent thermal efficiency while burning natural gas in a gas turbine exhausting to a conventional boiler-steam turbine system.[3]

The concept of generating steam in tubes immersed in fluidized beds of solids also is not new. Work has been under way for several years to develop this system, both in the United States and abroad. The British, in 1969, speculated that the advantages of pressurized fluid-bed boilers deserved further study including a mixed cycle incorporating a gas turbine.[4] In the United States, pilot-scale work has been under way for some four years to develop a system for pressurized fluid-bed combustion of solid wastes using hot exit gases to produce electric power in a gas turbine generator.[5]

The concept proposed herein includes elements from all of the aforementioned work; however, the cycle proposed is unique and has distinct advantages over other proposed or existing processes for processing wood waste and municipal refuse. The proposed cycle is shown in Figure 7; wood wastes would be dried to less than 10 percent moisture. This is important to the overall thermal efficiency because low-level turbine exhaust heat, much of which is wasted in other cycles, would be used to dry the wood wastes which may contain up to 50 percent moisture.

Thermodynamically, the cycle is very attractive, particularly for the combustion of waste materials with high water content. Performance data for one set of conditions, not necessarily optimum, are summarized in Table 1.

The plant can be divided into four major sections: feed preparation, fluid-bed burning, off-gas cleanup, and power generation facilities. In feed preparation, the removal efficiency of noncombustibles from the municipal refuse has a significant impact on the operation of the fluid bed. Operation of the fluid-bed burner (including feed introduction, ash removal, and in-bed heat transfer) and off-gas cleanup are critical to the successful operation of the proposed plant.

Table 1. PERFORMANCE DATA FOR PROPOSED CYCLE

Basic parameters	
Waste moisture content	50%
Gross heating value, dry	8075 Btu/lb
Air pressure to fluid bed	150 psia
Gas pressure to gas turbine	145 psia
Air temperature to compressor	60°F
Fluid bed and exit gas temperature	1440°F
Excess air	15%
Turbine exhaust pressure	17 psia
Turbine exhaust temperature	720°F
Steam pressure	1500 psia
Steam temperature	1000°F
Feed water temperature	415°F
Condensing pressure	1.5 in. Hg
Fluid-bed dryer and exit gas temperature	150°F
Calculated performance/100 lb dry fuel	
kWhr, steam turbo-generator	77.0
kWhr, gas turbine turbo-generator	11.5
Energy, kWhr/100 lb dry fuel	88.5
Thermal efficiency, %	37.5

The power generating facilities will be conventional and do not require detailed discussion. A process flowsheet of the demonstration plant is shown in Figure 8.

Feed Preparation and Storage

Feed to the fluid-bed burner consists of wood waste (e.g., sawdust, chips, and shredded material) and municipal refuse. Wood wastes are relatively homogeneous and require only sizing and drying before feeding to the bed. The heterogeneity of municipal refuse requires separation of glass and metals and sizing before being introduced to the fluidized bed. Based on a minimum amount of engineering development, the A-E would select a feed preparation scheme for facilitating materials handling, reducing environmental pollution, and providing safe storage.

Wood Waste Feed Preparation and Storage—Wood waste would be properly sized at the mill site for feed to the burner. Raw feed which is sufficiently dry (<10 percent moisture) would be transported directly to feed storage. All other feed would be dried approximately 10 percent moisture before storage.

Municipal Refuse Feed Preparation and Storage — With the exception of moisture content, a typical municipal refuse content and composition is shown in Table 2. The municipal refuse consist of glass, dirt, metal, and various combustible materials. Size distribution of the refuse varies from large pieces of material to dust particles; moisture content is a nominal 25 percent by weight. For rapid combustion, good quality fluidization, and satisfactory materials handling, refuse must be sized to less than 1-in. pieces. The water content is usually lowered to less than 10 percent during normal feed preparation (e.g., shredding and classification); therefore, drying of the refuse is not anticipated.

Table 2. TYPICAL MUNICIPAL REFUSE COMPOSITION

(Yard-free basis)

Material	Wt %
Paper, > 1/4 inch	48.9
Paper, wood, fabric fines	11.7
Wood	10.2
Fabrics	1.0
Plastics	2.0
Inerts (glass and metallics)	14.0
Dust	11.7
Heating value, Btu/lb (dry basis)	6000

A conceptual flowsheet for feed preparation of the municipal refuse is shown in Figure 9. The raw refuse is dumped into a feed-conveyor hopper directly from a truck; no raw feed storage is provided. After charging to the shredder, the material is transported to a classifier where the large and/or dense particles are separated in the underflow. As dictated by economics, ferrous metals may be separated from the underflow for recycle.

Feed preparation equipment will be oversized to allow for routine maintenance without limiting the plant capacity and would normally be operated only one shift per day. If economics require, refuse feed preparation could be done at the source; this is especially true in the case of a large city such as Spokane.

Fluid-Bed Combustion

The fluid-bed burner (Figure 10) is the heart of the proposed system. The maximum bed temperature is limited by the gas turbine blade materials and the minimum fusion temperature of the noncombustibles and ash present in the bed. Fluid-bed burners have a high heat release rate per unit volume of bed; because of the variability in the heat content of refuse, control of bed temperature could be difficult when charging municipal refuse alone. However, temperature control problems in the proposed concept should be minimal, since the wood waste (relatively homogeneous and of constant heat value) will comprise one-half to two-thirds of the feed to the burner.

Condensate within tubes of an in-bed heat exchanger is converted to steam by transfer of heat from the fluid bed. Depending on economics and technical considerations, bare or finned tubes will be used. Based on a practical carbon steel tube bundle configuration and overall heat transfer coefficients in the range of 300 to 600 Btu/hr-ft^2-°F, heat transfer rates in the range of 4×10^5 to 8×10^5 Btu/hr-ft^3 of bed are possible. For a 125 ton/hr plant, approximately 1×10^6 lb/hr of steam (1200 to 1500 psia and 900 to 1000°F) would be generated. Based on a nominal steam cycle efficiency of 35 percent, approximately 100 MW would be produced by the steam turbine.

Flue gases from the fluid-bed burner pass through a high efficiency off-gas cleanup system (series of cyclones followed by ceramic or sintered metal filters) for removal of essentially all particles >5 μm in diameter. The cleaned off-gas is then passed to a gas turbine where, based on a 125 ton/hr plant and a turbine cycle efficiency of 20 percent, 14 MW are produced.

Though most of the ferrous and nonferrous metals will be removed by magnetic separation and air classification, some of these components will remain in the feed to the burner. In addition, glass, dirt, ceramic material, and possible agglomerates of material (e.g., plastics, etc.) must be removed from the burner. If the material is allowed to accumulate on the air distributor, fluidization quality would deteriorate as a result of gas channeling. A reliable system for removal of ash agglomerate and noncombustibles is required.

Off-Gas Cleanup System

Removal of entrained particulates and corrosive gases is a major problem in the design of conventional incinerator off-gas cleanup systems; expansion of the off-gas in a turbine requires even a higher degree of off-gas cleanup. Particulates, if not removed, will erode and foul turbine blades and pollute the environment. Corrosive gases will also corrode the off-gas cleanup system and turbine blades. Fewer corrosive gases are generated during processing of wood waste; therefore, the wood waste could result in off-gas concentrations of corrosive gases of acceptable levels. Although maximum particulate loading specifications for turbine inlet gases vary, 2×10^{-3} gr/ft^3 of off-gas, with no more than 10 percent of the particles greater than 10 μm in size, is a typical requirement. Because of the control of bed temperature by transfer of heat to an in-bed steam generator, the excess air flow and hence volume of gas requiring cleanup is minimized using the proposed concept.

Conventional Particulate Removal — Particulates are normally removed from off-gas streams by combinations of cyclone separators, scrubbers, and electrostatic precipitators; each type of cleanup device has distinct disadvantages. Cyclones are normally effective in removing particulates at high gas rates as long as the particle size is larger than 20 μm. Scrubbers require low operating temperatures, and electrostatic precipitators are often ineffective. Sand or granular filters, though sometimes employed, are bulky and regeneration is difficult.

Combination Multi-Stage Cyclone and "Candle" Filters for Particulate Removal — The concept of filtering off-gases at high temperatures is not new; sintered metal filters are commonly employed up to 1500°F. Such filters are available for removing submicron particles at pressure differentials less than 1-in. water. Ceramic "candle" filters, used in conjunction with multi-stage cyclones, appear to be most attractive for removing particulates to the levels recommended by turbine manufacturers. Well designed 3-stage cyclones could almost satisfy the requirements alone, and candle filters could further reduce particulate loading to levels below process requirements. The cyclones would be constructed of a material resistant to chloride corrosion at temperatures as high as 1500°F.

In the proposed concept, off-gases at 145 psia and 1400 to 1500°F would leave the burner and pass through multi-stage cyclones followed by candle filters for final particulate removal, as shown in Figure 11. The multi-stage cyclones would remove essentially all particulates greater than 20 μm. Off-gas passing to the turbine would contain a maximum loading of 1×10^{-3} gr/ft^3, and essentially all particulates greater than 5 microns would be removed. The multi-stage cyclone and candle filter systems are commercially available.

CONCLUSIONS

Disposal of municipal refuse and wood waste while minimizing harmful effects on the environment is an existing problem. The per capita increase of refuse generation rates coupled with the expected population increase will almost double the annual refuse generated by 1980. Wood waste generation rates are also expected to increase, though not as rapidly as municipal refuse.

Existing methods for municipal refuse disposal (landfill and conventional incineration) frequently result in air and water pollution and unsightly facilities. Disposal of wood waste by incineration in teepee burners is unacceptable from the standpoint of satisfying air pollution standards. Development of more advanced systems for disposal of municipal refuse is based largely on the concept of product recycle. The recycled products have questionable market value when compared with virgin materials.

Existing and near-term fluid-bed technology can result in a fluid-bed process for power generation having a thermal efficiency greater than 35 percent while reducing the off-gas mass flow rates per MW-hour of electrical energy by a factor of eight when compared with a conventional gas turbine cycle. In such a process, steam would be generated within an in-bed heat exchanger and used as the working fluid in a steam-turbine cycle. The latest technology in finned-tube heat transfer within fluid beds would be used. The burner off-gases would be cleaned of particulate by passage through a high efficiency cleanup system consisting of staged cyclones and sintered metal (stainless steel and Hastelloy C) or ceramic "candle" filters. Particulate removal at the temperatures (1400 to 1500°F) proposed for refuse and wood waste combustion has been tested in development and operations facilities in the atomic energy field. Use of existing technology and commercially available equipment where applicable will significantly reduce the development costs and time required to place a full-scale waste disposal-power generation facility on stream. Power production costs are estimated to be in the range of 5 to 6 mills/kWhr.

BIBLIOGRAPHY

1. Bendixsen, C.L. et al. The Third Processing Campaign in the Waste Calcining Facility. U.S. Atomic Energy Commission, Oak ridge, Tenn. Publication Number IN-1474. May 1971.
2. Kilian, D.C. et al. Description of the Pilot Plant for the Headend Reprocessing of Unirradiated Rover Fuels. U.S. Atomic Energy Commission, Oak Ridge, Tenn. Publication Number IN-1181. May 1968.

3. Cox, A.R., et al. Operation of San Angelo Power Station Combined Steam and Gas Turbine Cycles. In: Proceedings American Power Conference, Volume XXIX, p. 401-412.

4. Big Economics from Pressurized Fluid-Bed Combustion. The Engineer, July 24, 1969.

5. Combustion Power Unit-400. Combustion Power Co., Palo Alto, Calif. Prepared for the Bureau of Solid Waste Management, Environmental Protection Agency, Washington, D.C. under Contract Number Ph 86-67-259.

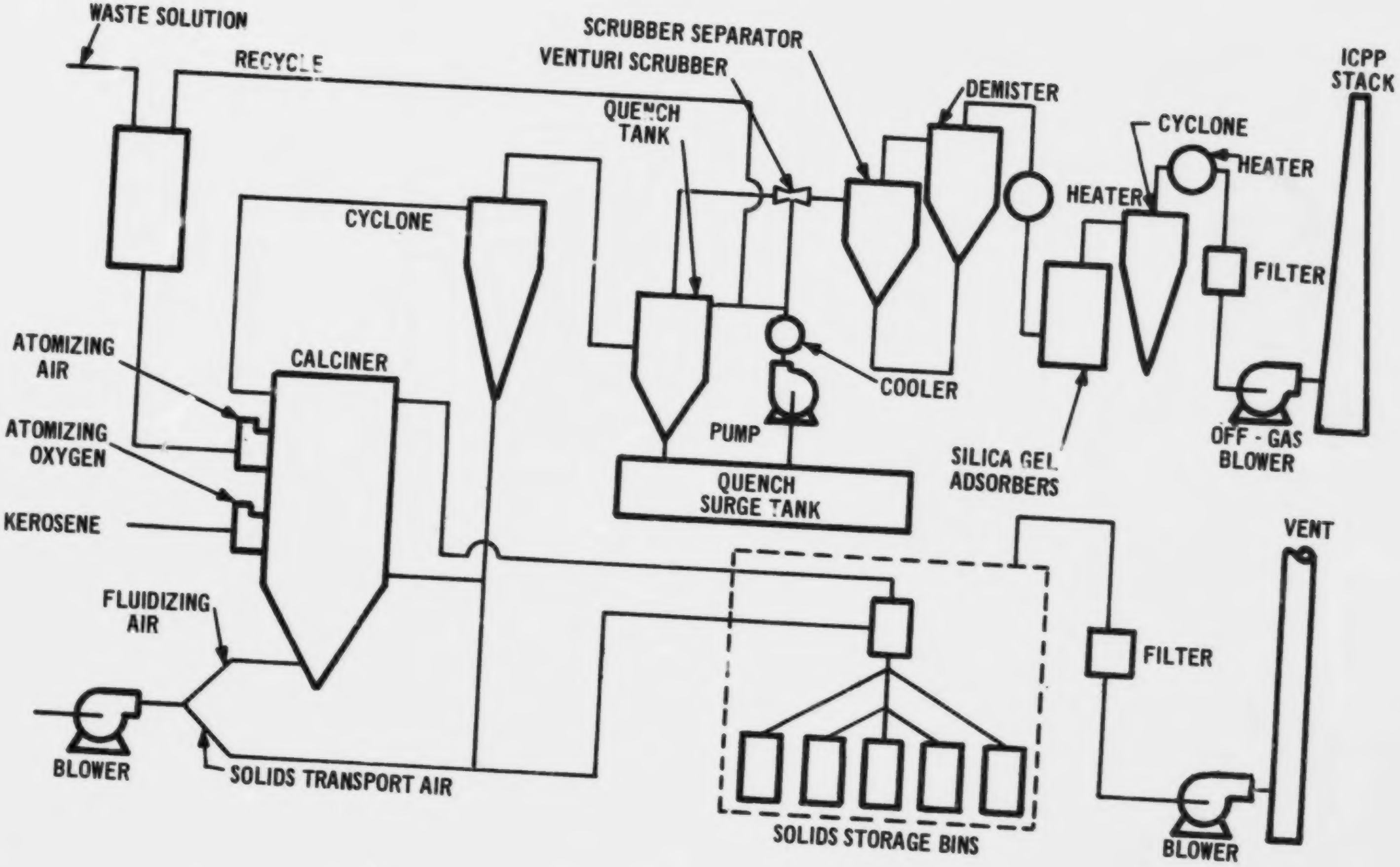


Figure 1. Waste calcinating facility.

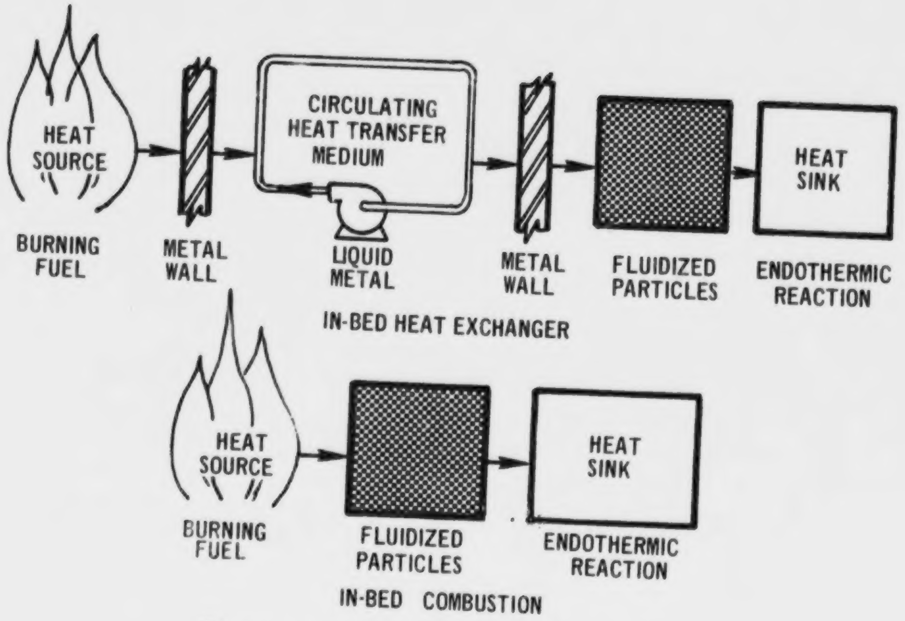


Figure 2. Comparison of heat flow paths.

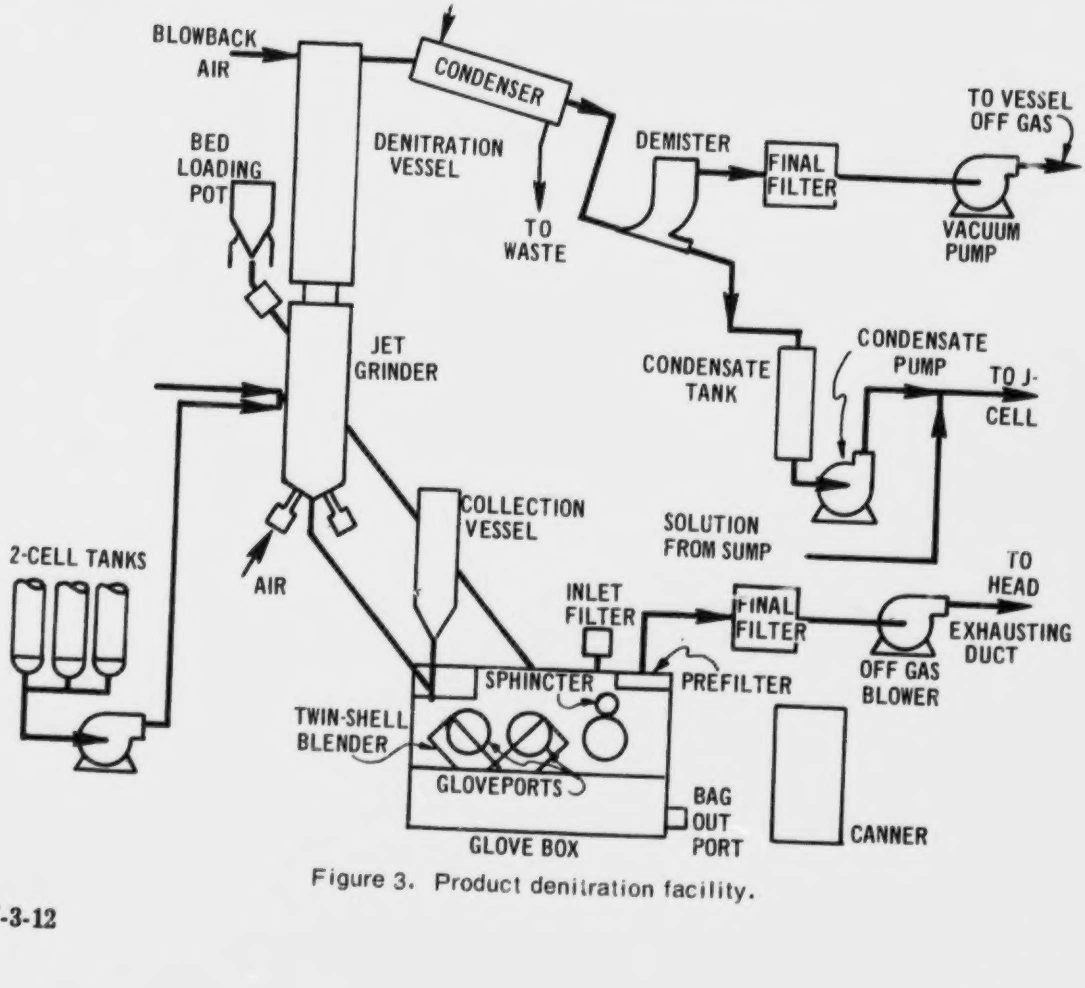


Figure 3. Product denitration facility.

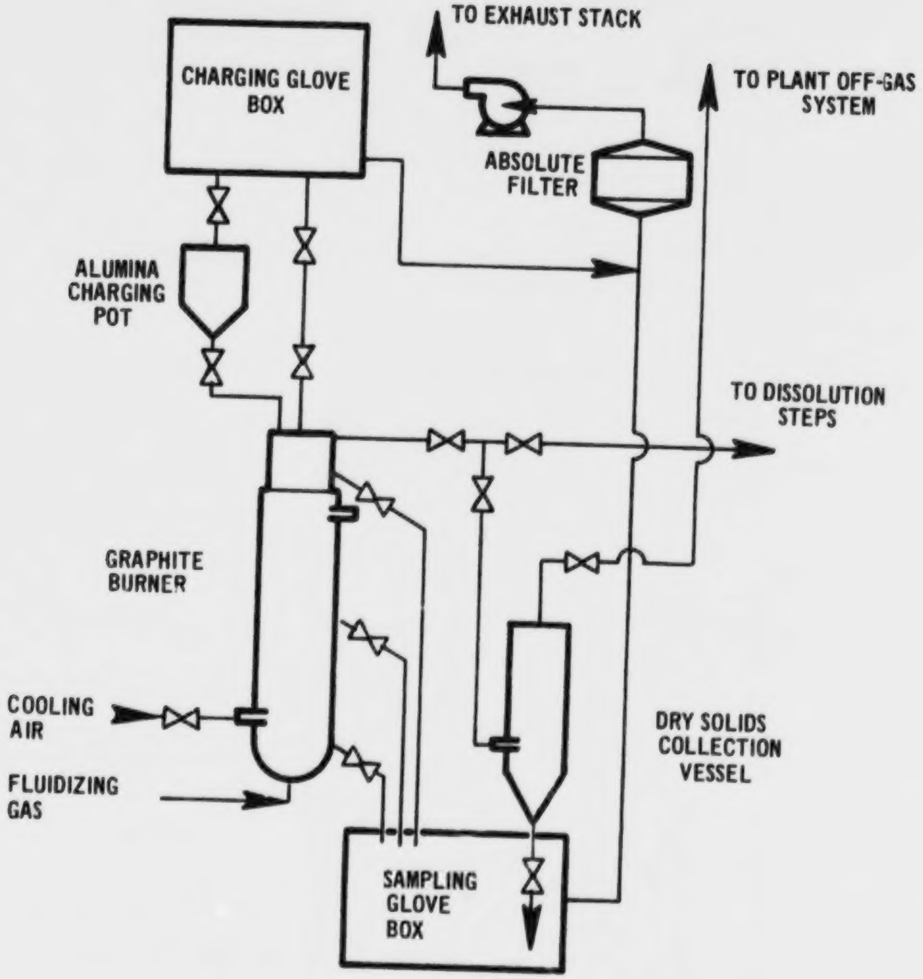


Figure 4. Graphite fuels pilot plant-combustion process flowsheet.

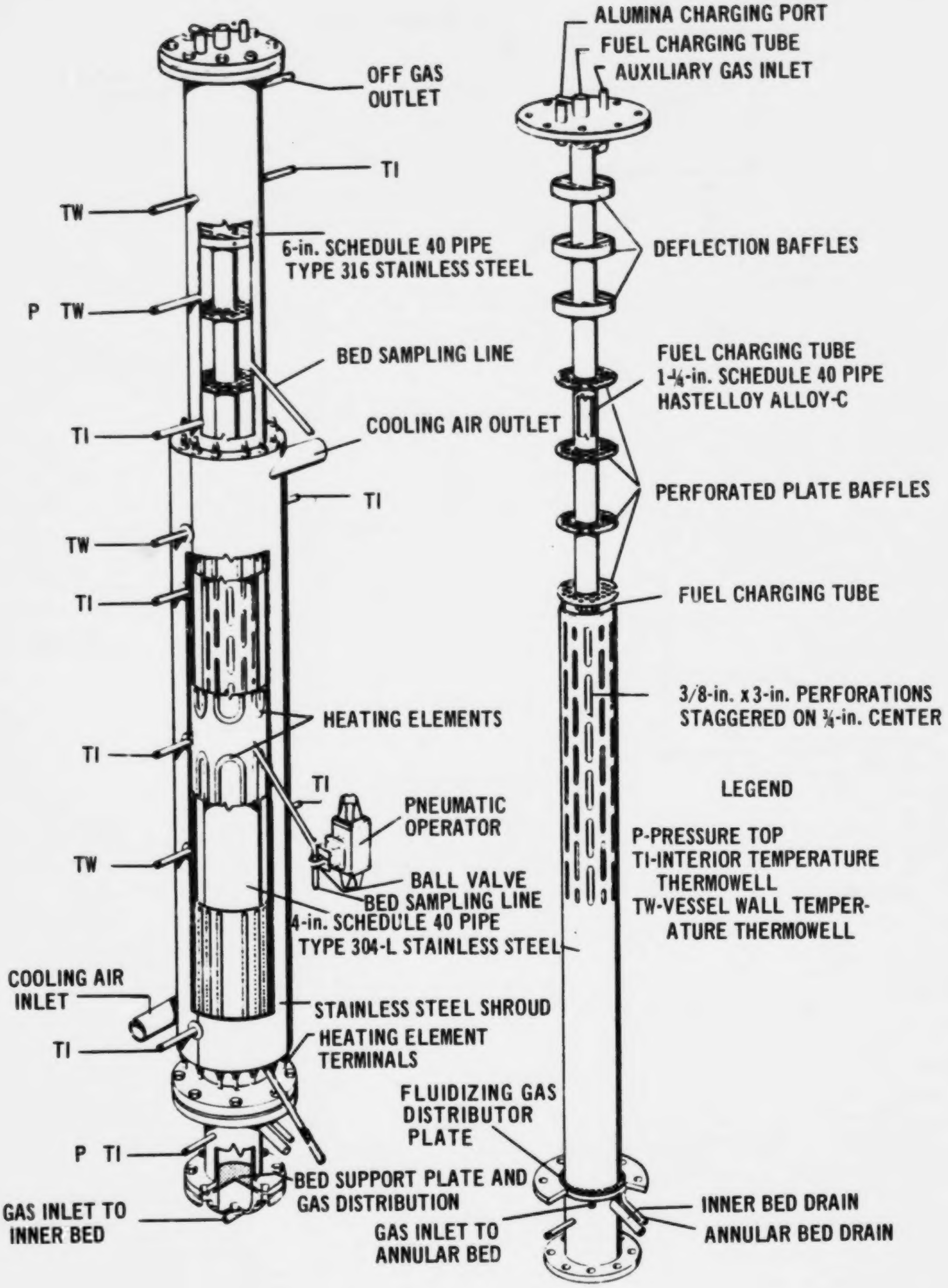


Figure 5. Concentric bed burner.

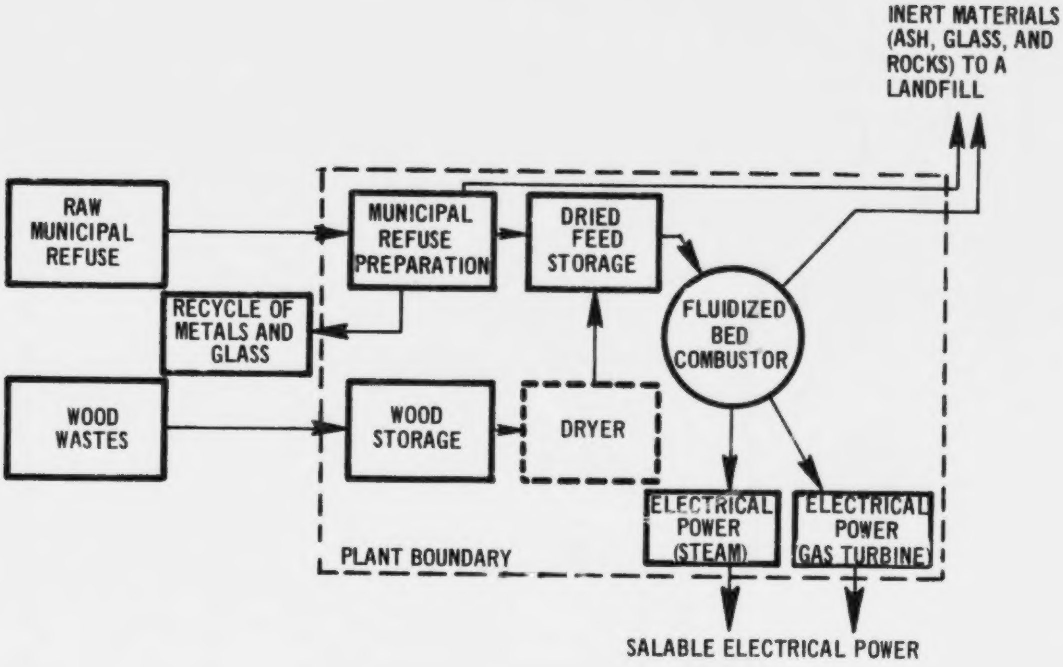


Figure 6. Schematic of waste disposal-power generation facility.

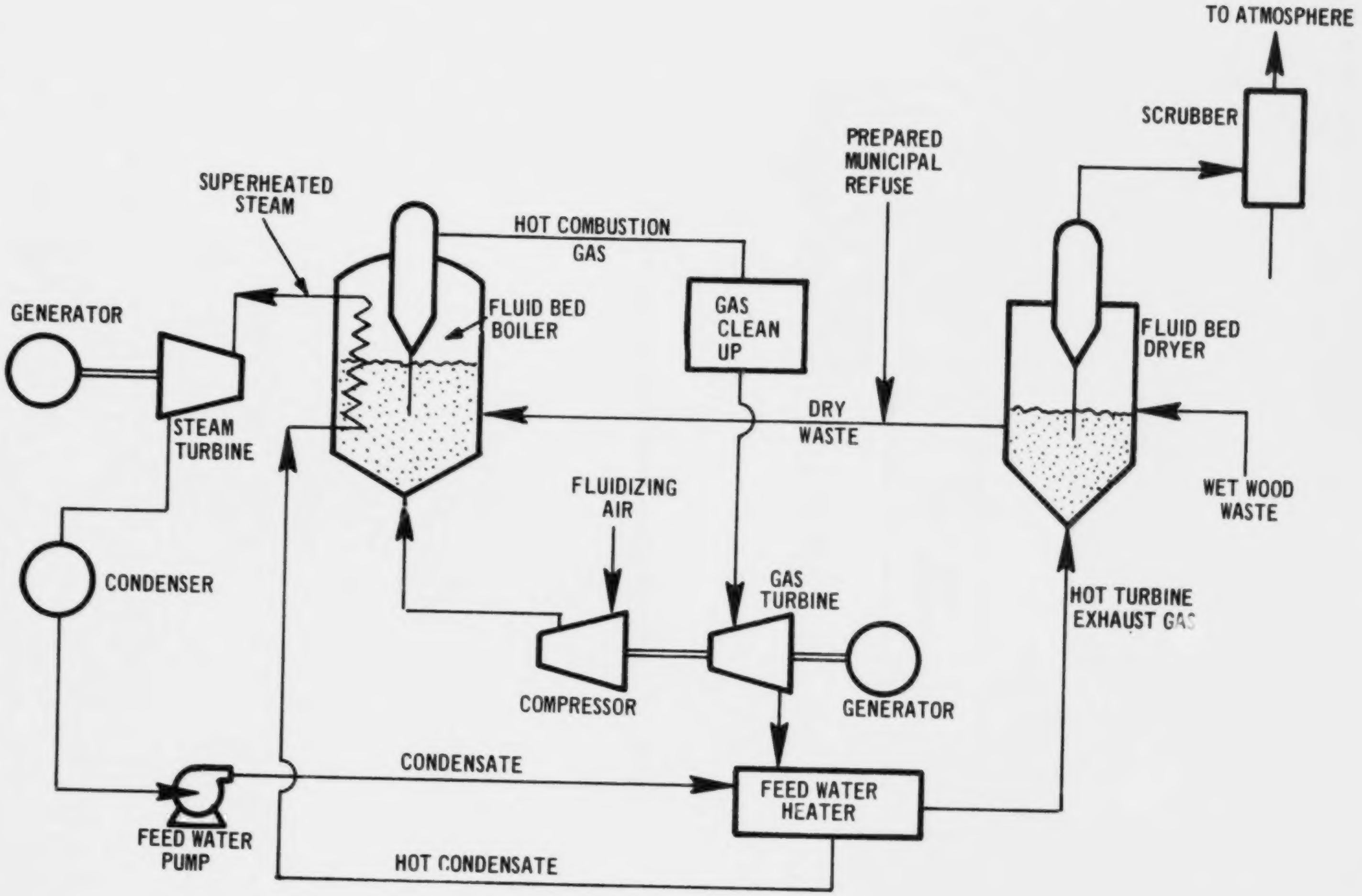


Figure 7. Proposed cycle for producing electric power from wood waste and municipal refuse.

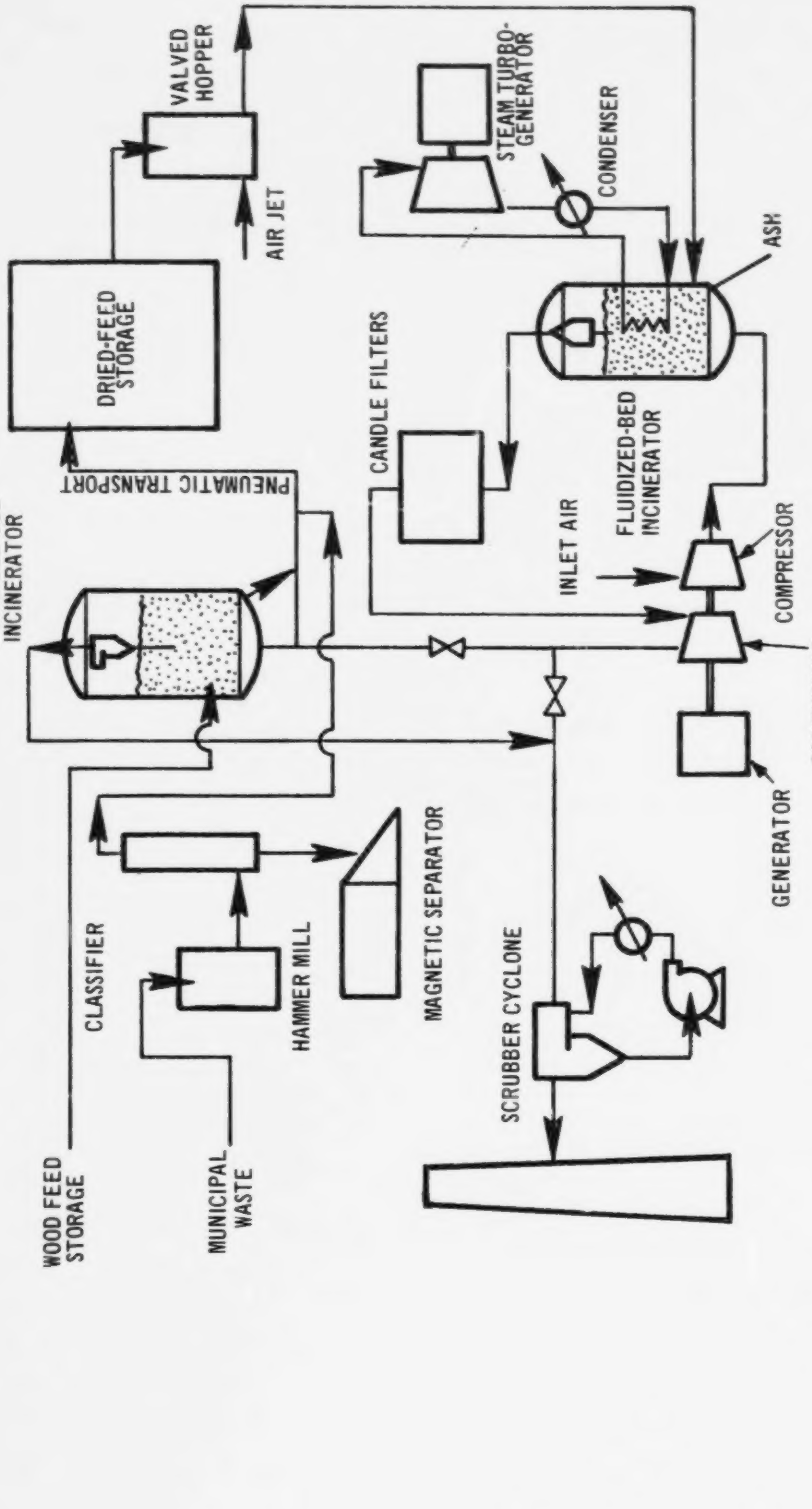


Figure 8. Flowsheet of proposed waste recycle-power plant.

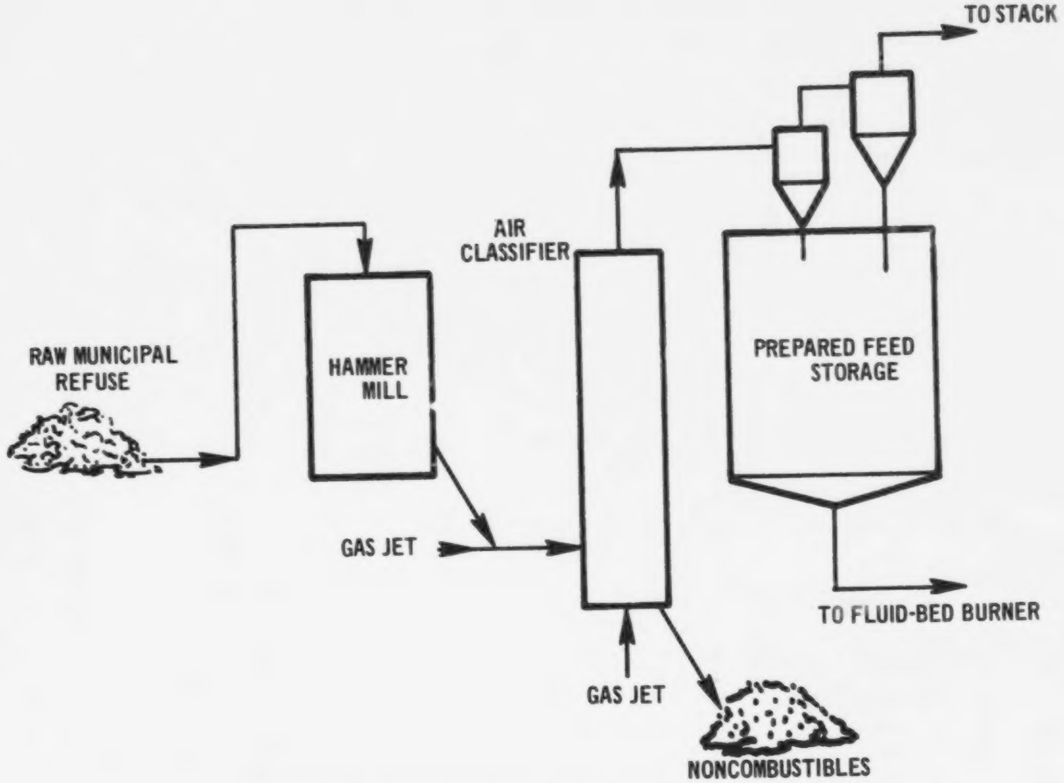


Figure 9. Municipal refuse feed preparation facility.

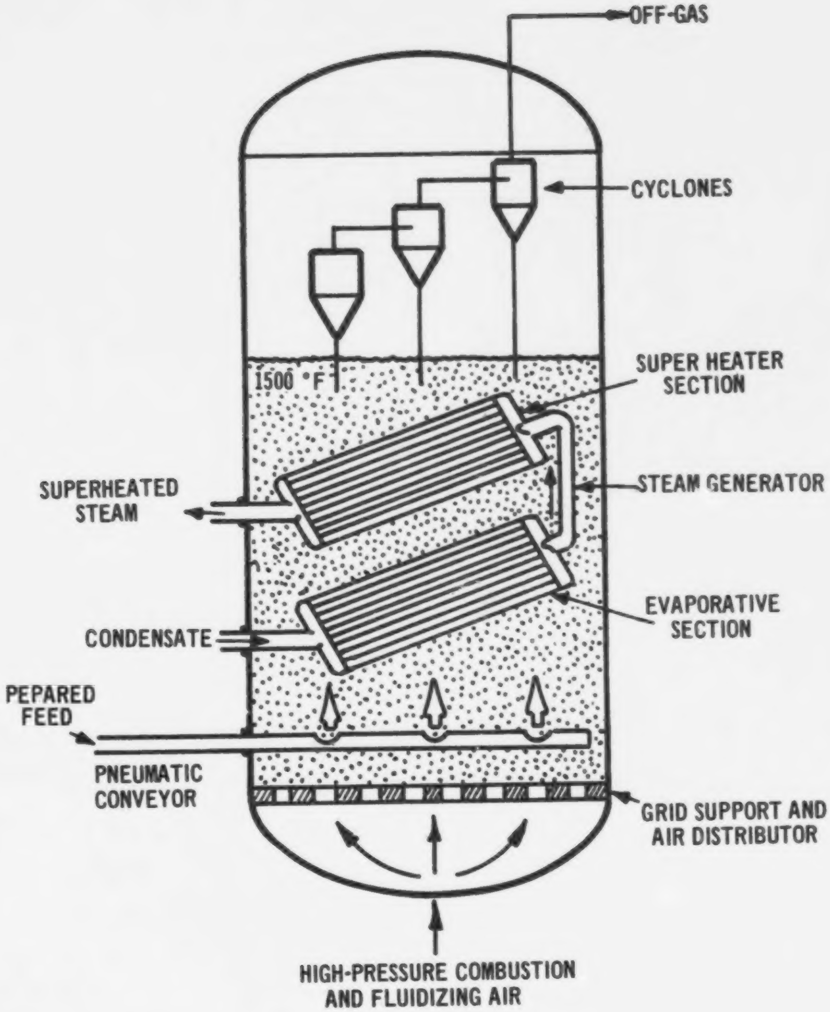


Figure 10. Fluid-bed burner.

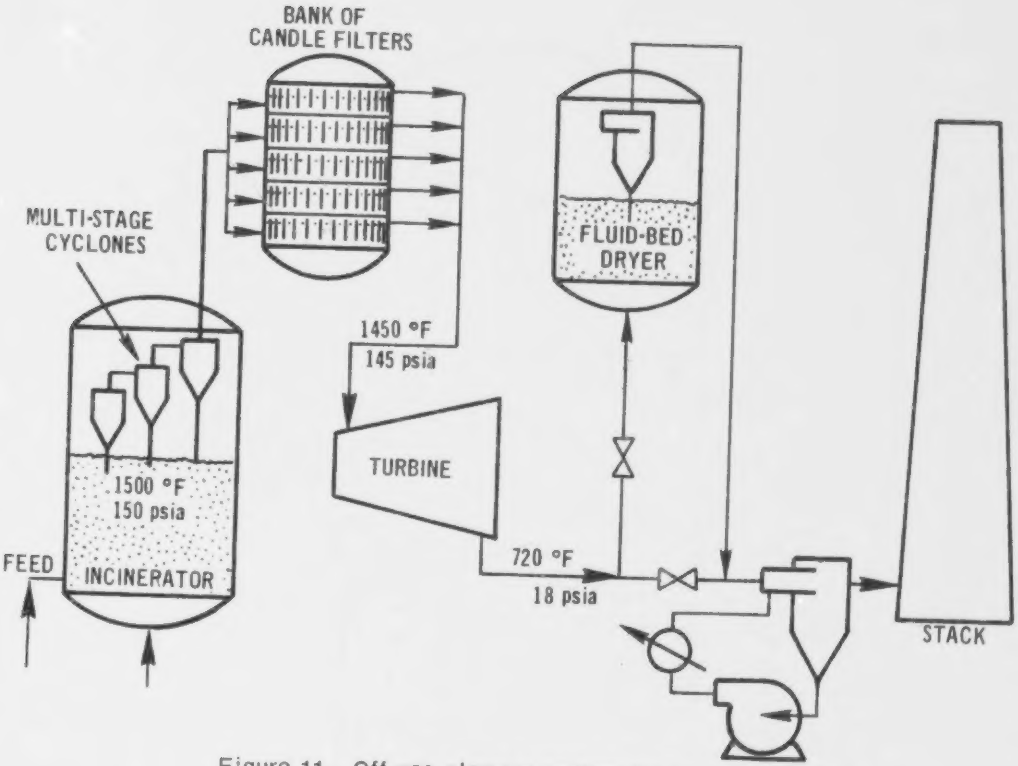


Figure 11. Off-gas cleanup system for proposed plant.

4. POWER GENERATION USING THE SHELL GASIFICATION PROCESS

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ABSTRACT

Growing concern about sulfur and nitrogen oxide emissions has given rise to a search for means of converting conventional fuels into clean, non-polluting fuels for electric power generation. Through its ability for converting liquid fuels into partially oxidized gaseous fuels and recovering the heat of partial oxidation in the form of high pressure steam, the Shell Gasification Process (SGP), aided by the Shell Sulfinol or ADIP Process, offers an attractive means of converting sulfur-laden heavy hydrocarbon feedstocks of high metals content into non-polluting fuel gas and saleable elemental sulfur.

Conceptual design and economics of an SGP-based power plant utilizing the Combined Gas and Steam (COGAS) cycle are presented. Besides offering the simplicity, flexibility, and reliability associated with the SGP, such a power plant can generate electric power at unit costs competitive with those of future conventional power plants.

INTRODUCTION

The Shell Gasification Process (SGP)* is a process for the partial combustion of hydrocarbons, and is particularly suitable for the partial combustion of heavy, sulfur containing residual fuels and heavy crude oils to produce a mixture of hydrogen and carbon monoxide. From this mixture the hydrogen sulfide produced during partial oxidation can be readily removed. A non-polluting fuel gas is thus produced which can be used for power generation. This type of fuel should be of particular interest for power generation because of the following factors:

1. Natural gas, for many years a sulfur-free fuel, has slid into a declining reserve position in the face of an increasing demand at present regulated prices.
2. The cost and difficulty of desulfurizing heavy fuel oils, particularly those with high metals content, is very high.
3. Eastern and midwestern coals have high sulfur contents which to an increasing extent make them unsuitable for use for generation of power in conventional steam power plants.
4. There is a relatively long lead time for the development of low sulfur western coals, and also high transportation cost associated with the use of these coals.
5. There is a long lead time required for the installation of nuclear power plants.

As an alternative method of power generation, the Shell Gasification Process, with a moderate investment and a high thermal efficiency (>85 percent), converts fuels with high levels of sulfur, nitrogen and/or metals into attractive power generating fuels for use in the COGAS cycle (Combined Gas and Steam cycle).

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*Licensed by Shell Development Company, Houston, Texas 77001 and Shell Internationale Research, Maatschappij, N.V., The Hague.

An SGP-based power plant consists of a Shell Gasification Process unit, for converting residual fuels or low-value crudes into low-Btu fuel gas and recovered steam, and a gas turbine-steam turbine unit for converting these products into electrical power. An SGP-based power plant differs from a conventional thermal power plant: in a conventional thermal power plant raw fuel is burned directly, while in an SGP-based plant the power plant fuel is the product of partial oxidation of the raw fuel.

The power generation unit recommended for the SGP-based power plant uses the COGAS cycle. The COGAS cycle[1,2,3] is thermodynamically superior to either the steam cycle or the gas cycle. It is particularly suited for an SGP-based power station, since in the SGP the net exothermic heat of partial oxidation is recovered as high pressure steam which can be integrated with the steam section of the COGAS cycle.

The following technical and economic case study shows that an SGP-based power station is not only a feasible means of generating power without contributing to atmospheric pollution, but it is also economically competitive with conventional power plants of the future.

CHEMISTRY OF PARTIAL OXIDATION

Partial oxidation describes the net effect of a number of component reactions that occur in a flame, supplied with less than stoichiometric oxygen. This net effect can be approximated:

$$C_nH_m + (\frac{n}{2})O_2 \rightarrow n\,CO + (\frac{m}{2})H_2 \quad (1)$$

and is actually a combination of several reactions that occur within the reactor.

Heating-up and cracking phase

In the fuel injection region of the reactor, hydrocarbons leaving the atomizer at about preheat temperature are intimately mixed with air. Prior to combustion they are heated and vaporized by back radiation from the flame and the reactor walls. Some cracking of the hydrocarbons to carbon, methane, and hydrocarbon radicals may take place during this brief interval.

Reaction phase

As soon as the ignition temperature is reached, part of the hydrocarbons will react with oxygen according to the highly exothermic reaction:

$$C_nH_m + (n + \frac{m}{4})O_2 \rightarrow n\,CO_2 + \frac{m}{2}H_2O. \quad (2)$$

As the equilibrium is far to the right, practically all the available oxygen is consumed in this phase. The remaining hydrocarbons which have not been oxidized react with steam and the combustion products from reaction (2) according to the endothermic reactions:

$$C_nH_m + n\,CO_2 \rightarrow 2n\,CO + \frac{m}{2}H_2 \quad (3)$$

and

$$C_nH_m + n\,H_2O \rightarrow n\,CO + (\frac{m}{2} + n)H_2. \quad (4)$$

In order to prevent excessive local temperatures, it is essential that all reactants of equations (2) to (4) are intimately mixed so that the endothermic reactions tend to balance the exothermic reactions. In this way the complex of reactions is brought to a thermal equilibrium resulting in a measured temperature of about 2350 to 2550°F.

Soaking phase

Soaking takes place in the rest of the reactor where the gas is at a high temperature. The gas composition changes only slightly due to secondary reactions of methane and carbon and the water gas shift reaction.

Methane produced by cracking will decrease according to:

$$CH_4 + H_2O \rightleftharpoons CO + 3H_2 \quad (5)$$

and

$$CH_4 + CO_2 \rightleftharpoons 2CO + 2H_2. \quad (6)$$

As the reaction rate is relatively low, the methane content is higher than would be expected from equilibrium.

During the soaking phase a portion of the carbon also disappears according to the reactions:

$$C + CO_2 \rightleftharpoons 2CO \quad (7)$$

and

$$C + H_2O \rightleftharpoons CO + H_2. \quad (8)$$

However, some carbon is always present in the product gas from the reactor in a quantity equivalent to about 3 weight percent of the oil feed.

The composition of the fuel gas is determined by the water-gas shift equilibrium which appears to freeze after the gas enters the waste heat boiler at an equilibrium temperature about 2200 to 2400°F.

$$CO + H_2O \rightleftharpoons CO_2 + H_2 \quad (9)$$

DESCRIPTION OF SHELL GASIFICATION PROCESS

A simplified SGP flow sheet is given in Figure 1. The hydrocarbon charge and the oxidant are preheated and fed to the reactor.

The hot reactor-effluent gas containing about 3 percent of the feed as soot is passed to a waste heat boiler, producing high pressure saturated steam. High heat transfer rates are achieved, with the result that the temperature of the gas leaving the waste heat boiler closely approaches that of the steam produced in the boiler. The design and construction of the waste heat boiler are such that the surface remains clean for an indefinite period (without using any external cleaning devices); it may be noted that the waste heat boiler of the Shell prototype unit has been in operation since 1956 and never has been cleaned on the gas side. The waste heat boiler can be designed for steam pressures up to about 1500 psig.

The crude gas leaving the waste heat boiler at temperatures around 350°F is then passed to the carbon removal system, consisting of a bulk removal of the carbon by a special method of contacting the gas with water, and a final water wash. The product gas is virtually free of carbon (<5 ppm).

The carbon produced in the gasification is recovered as a soot-in-water slurry (carbon content 1 to 2 weight percent). In most cases, it will not be possible to dispose of this carbon slurry as such. Therefore, a special technique has been developed for removing the carbon from the slurry, resulting in carbon-free water for re-use. Depending upon the metals content of the feedstock and the economics and maintenance policy of the process operator, up to 100 percent of the soot can be recycled to extinction with the fresh feed.

Sulfur in the feedstock is converted primarily to H_2S and traces of COS. The carbon-free product gas is treated in a Shell Sulfinol or ADIP process unit where the sulfur compounds and most of the CO_2 are absorbed. The desulfurized gas typically contains less than 5 ppm of sulfur. The acid gas effluent from the Sulfinol unit is fed to a Claus process unit which recovers elemental, salable sulfur.

Depending on the desired LHV (Lower Heating Value) in the product gas, either oxygen or air (enriched or unenriched) may be used as the oxidant. Nitrogen present in the air acts as a moderator for temperature control in the reactor. When either oxygen or air enriched with oxygen is used as the oxidant, a certain quantity of steam must be injected into the reactor for temperature moderation. Air oxidation produces a low heating value (120 Btu/scf) fuel gas due to the presence of nitrogen, while oxygen feed produces a medium heating value gas (300 Btu/scf). Typical product gas compositions for air and oxygen gasification are shown in Table 1.

Table 1. TYPICAL PRODUCT GAS COMPOSITION

	% vol, dry basis	
	Oxygen oxidation	Air oxidation
Hydrogen	48.0	12.0
Carbon monoxide	51.0	21.0
Methane	0.6	0.6
Nitrogen	0.2	66.0
Argon	0.2	0.4
Sulfur	5 ppm	5 ppm
Total	100.0	100.0

COGAS CYCLE THERMODYNAMICS

Although the idea of combining a gas turbine and steam turbine is old, its application to power generation has been studied only lately. Wood[3] has presented an excellent summary of the development of the COGAS cycle. In a COGAS cycle (Figure 2), air is compressed and heated by burning fuel in it. The hot gases are then expanded in a gas turbine coupled to the air compressor and a generator. The gas turbine exhaust, still at a high temperature, is used to raise and superheat high pressure steam; it is also used as a heat source for deaeration and boiler feed water preheating. The steam generated by the gas turbine exhaust is expanded in a steam turbine to produce additional electric power. Heat rejection occurs in the stack exhaust and in the condenser of the steam cycle. It is well known that the greater the difference between the heat source and heat sink temperatures of any heat engine, the higher its thermodynamic efficiency. In the COGAS cycle, the heat sink of the gas cycle becomes the heat source of the steam cycle, increasing the overall spread between source and sink temperatures for the combined cycle. As a result, the COGAS cycle has a higher thermodynamic efficiency than either the simple gas cycle or the steam cycle. In the case presented in this paper, a COGAS cycle efficiency (based on the net useful energy input to the power plant) of 44 percent was obtained. Simple gas cycle and steam cycle efficiencies are of the order of 25 percent and 37 percent, respectively.

In application of the Shell Gasification Process to the COGAS cycle, the addition of the steam generated by the heat of partial oxidation to the steam generated in the COGAS cycle largely compensates for the loss of heating value of the oil caused by gasification.

POWER PLANT FLOW SCHEME

A flow diagram of the power plant using desulfurized fuel gas as fuel is shown in Figure 3. Air is compressed to 14 atm and split into two parts. One part is cooled by heat exchange with 100°F product gas from the SGP section and compressed again in a booster compressor to 18 atm before entering the SGP unit as the oxidant. The other part of the compressed air is combusted to a temperature of about 2200°F in the gas turbine combustor by burning the sulfur-free fuel gas supplied by the SGP/Sulfinol units, and expanded to 1.5 atm (absolute) pressure in the gas turbine which is coupled to an electric power generator. The gas turbine exhaust is cooled to about 350°F in a waste heat boiler/superheater and boiler feed water deaerator before being vented to atmosphere. Steam (1250 psig) generated in the SGP waste heat boiler is combined with the steam generated in the power plant waste heat boiler, and the combined steam is superheated to 1000°F in the superheater section of the latter. The superheated steam is expanded typically to about 4-in. Hg vacuum in a steam turbine coupled to a second electric power generator. From the point of view of startup and control, it is advisable to use separate generators for the gas and steam turbines. Approximately 56 percent of the total power generation is contributed by the gas turbine.

SGP-BASED POWER PLANT — CASE STUDY

Using the foregoing technology, an economic study has been made of an SGP-based power plant of 200-MW nominal generating capacity. Air was used as the oxidant in the partial oxidation step. The composition of the typical heavy residue used as feedstock is stated in Table 2. The material balance of various gas streams in the power plant appears in the inset of Figure 3.

Table 2. TYPICAL RESIDUAL FEED PROPERTIES

Gravity, °API, 60°F	12.0	
Specific gravity, 60/60	0.986	
Composition, wt %		
Carbon	86.00	
Hydrogen	10.73	
Sulfur	2.65	
Nitrogen	0.30	
Oxygen	0.30	
Ash	0.02	
Total	100.00	
Viscosity, centistokes		
470°F	4	
212°F	55	
100°F	800	
Ash analysis:	ppm in feed	% of ash
Nickel	30	15.0
Vanadium	100	50.0
Sodium	1	0.5
Iron	4	2.0
Others	65	32.5
Total	200	100.0

Turbine manufacturers have indicated to us that industrial gas turbines are currently designed for a compression ratio of 12 and turbine inlet temperature of 1800°F. However, it is predicted that by 1975, these conditions are likely to be upgraded to 14 and 2200°F, respectively. In this evaluation we have assumed 1975 technology. Other simplifying assumptions made were:

1. The gas turbine can accommodate an increase of approximately 25 percent (mole) in the gas flow across the combustor as low-Btu gas is injected into the combustor. (This has been shown to be feasible.)
2. Polytropic efficiencies of the various components are : compressor 90 percent, gas turbine 90 percent, steam turbine 75 percent[2].
3. Approximately 2 percent of boiler feed water is evaporated in the deaerator, and 5 percent is rejected in the boiler feed water blowdown. Thus, fresh boiler feed water requirement is 7 percent of the total steam generation.
4. Friction and generator losses, which are normally small, have been neglected.

RESULTS AND DISCUSSION

A power flow diagram for this case (Figures 4A and 4B) shows the factors contributing to an overall station efficiency of 38 percent. Figure 4A refers to the SGP unit and 4B shows the power flow through the power generation unit. The downstream end of Figure 4A thus matches with the upstream end of Figure 4B. About 87 percent of the energy input to the SGP unit (LHV basis) is available to the power plant as input energy.

Table 3 is a summary of the capital and manufacturing costs. The major uncertainties in the unit power cost in this estimate, the power plant capital cost and the oil price, are shown parametrically (Figure 5) based on 1972 U.S. East Coast costs.

Estimated unit power costs are about 1 to 2 mills/kWhr higher than current rates, depending on the location. However, the latter costs do not include the cost of stack gas scrubbing or other alternatives of treating sulfur and NO_x emissions. With the growing shortage of natural gas and restrictions on sulfur, NO_x, and particulate emissions, the power cost is certain to rise rapidly in the next few years. At such time, SGP-based power plants will offer an attractive alternative to conventional power plants.

Some of the advantages of the Shell Gasification Process relative to coal gasification processes are:

1. The SGP unit flexibly accepts a wide variety of fuels ranging from heavy residues (e.g., flasher pitch) to natural gas. Thus, a consistent and continuous quality of fuel gas can be generated despite variations in the quality of fuel supply.
2. The SGP-based power station handles fluids, avoiding the complex solids handling andash disposal steps involved in coal gasification units.
3. Both SGP and coal gasification units require comparable installation times. The lag in coal availability may lead to a longer project realization time for coal gasification-power units.

Relative to other means of clean power generation, such as the conventional power plants with stack gas clean-up units, the SGP-based power plant offers the following advantages:

1. The SGP unit, followed by a Sulfinol and Claus unit, produces the most marketable quality of recovered sulfur.
2. The NO_x concentration in SGP-based fuel gas is low because the C-N bond in the fuel is broken mostly into CO and N_2. With careful gas turbine combustor design there is little breaking of the N-N bond, to produce NO_x.

The unit evaluated in this paper is intended for intermediate to base load applications.

Table 3. POWER GENERATION COST

Power produced (gross), MW[a]	200.0
Power consumed, MW	4.7
Net power output, MW	195.3
Overall efficiency, %	38.0
Capital costs, $ x 10^6	
Fuel processing unit	18.2
Power generation unit	31.4
Total capital cost, $ x 10^6 [b]	49.6

Operating cost	$ x 10^6/yr	Mills/kWhr
Variable costs		
Oil @ $x/bbl	2.61x	1.52x
Sulfur credit @ $10/ton	(0.11)	(0.06)
Catalysts and chemicals	0.10	0.06
Cooling and boiler feed water[c]	0.69	0.40
Total	0.68 + 2.61x	0.40 + 1.52x
Fixed costs		
Operating labor @ $83,500/job (4 operators)	0.34	0.20
Maintenance @ 3% of capital	1.45	0.85
Local overhead @ 100% labor plus 25% maintenance	0.70	0.41
Taxes and insurance @ 1% of capital	0.50	0.29
Total	2.99	1.75
Net operating cost	3.67 + 2.61x	2.15 + 1.52x
Capital charges @ 14%	6.94	4.06
Total power cost	10.61 + 2.61x	6.21 + 1.52x

[a] Yearly average value. Actual capacity is 11% higher to compensate for 90% stream factor.

[b] 1972, U.S. East Coast dollars.

[c] C. W. at 1.5 ¢/10^3gal.
Cond. at 7% of circulation @ $0.50/$10^3$ gal.

Basis: 200 MW Nominal Generating Capacity
90% Stream Factor

CONCLUSIONS

The Shell Gasification Process in conjunction with a COGAS power generation unit offers an attractive combination for generating electric power from high-sulfur, heavy residual fuels. At the same time, sulfur and NO_x emissions are almost completely eliminated, and the sulfur is recovered as a salable byproduct. The process flexibly accepts wide and frequent variations in the feed quality and composition ranging from natural gas to flasher pitch. Effluents are minimized. Efficient heat recovery in the gasification unit combined with the advantages of the COGAS cycle in the power generation unit ensures very little loss of energy despite the additional fuel processing. High stream factors render this process suitable for base load applications. In view of these merits, the SGP-based power station has promise for use in the electric utility industry.

REFERENCES

1. Business Week, March 11, 1972, p. 44C.

2. Robson, R. L., et al. Technological and Economic Feasibility of Advanced Power Cycles and Methods of Producing Nonpolluting Fuels for Utility Power Stations. United Aircraft Research Laboratories, East Hartford, Connecticut. Report Number J-970855-13, p. 258.

3. Wood, B. Combined Cycles: A General Review of Achievements. Combustion, April 1972, p. 12.

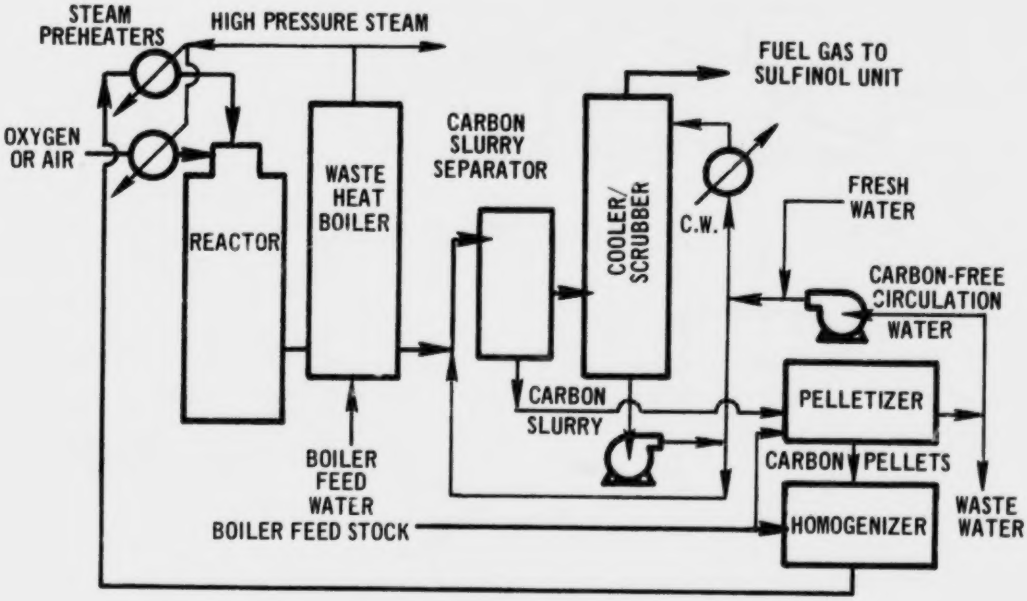


Figure 1. SGP for fuel gas manufacture.

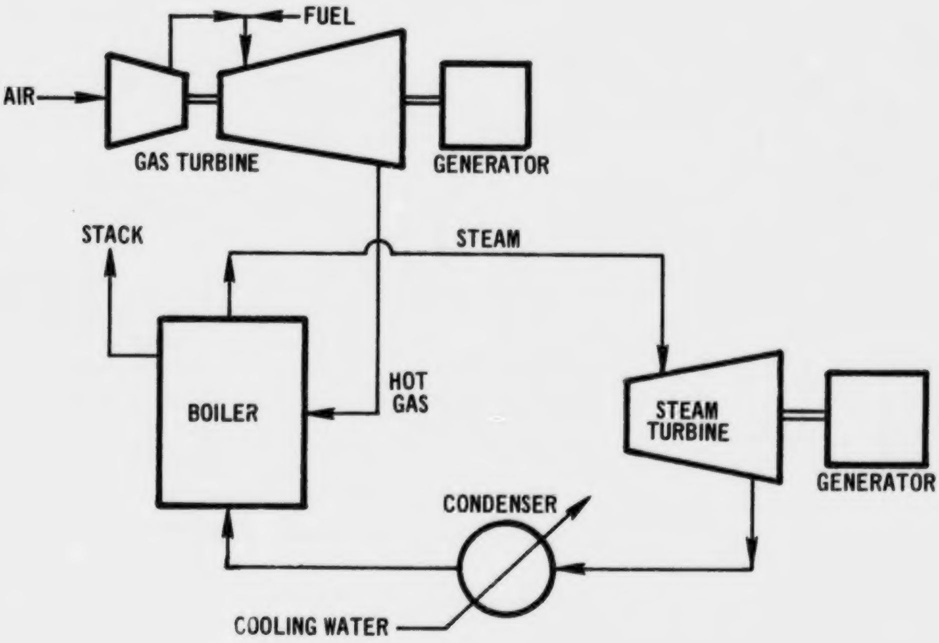
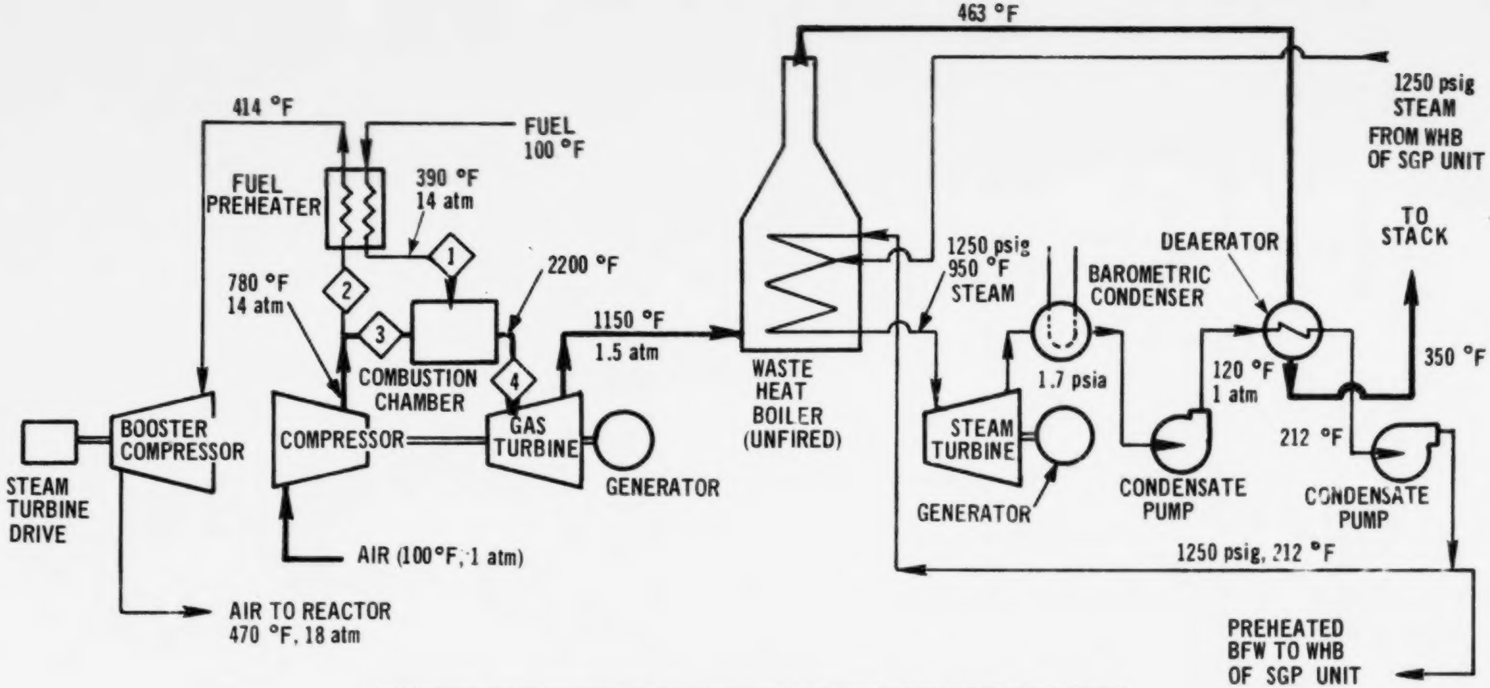


Figure 2. COGAS cycle for power generation.



COMPONENT	MOL WT	B.P.	SP. GR.	ALL NUMBERS IN lb-mole hr			
				CASE 1		116.5 Btu/scf	
				1	2	3	4
HYDROGEN	2.0			4,158			
CARBON MONOXIDE	28.0			6,490			
CARBON DIOXIDE	44.0			397			6,972
METHANE	16.0			85			
NITROGEN	28.0			17,230	17,485	64,412	81,642
OXYGEN	32.0				4,647	17,112	11,668
WATER	18.0						4,308
TOTAL				28,540	22,150	81,534	104,560
PRESSURE AND TEMPERATURE, °F				14 atm, 780°F			2,200°F
scfm x 10^3				179	140	516	661

Figure 3. Power plant flow diagram.

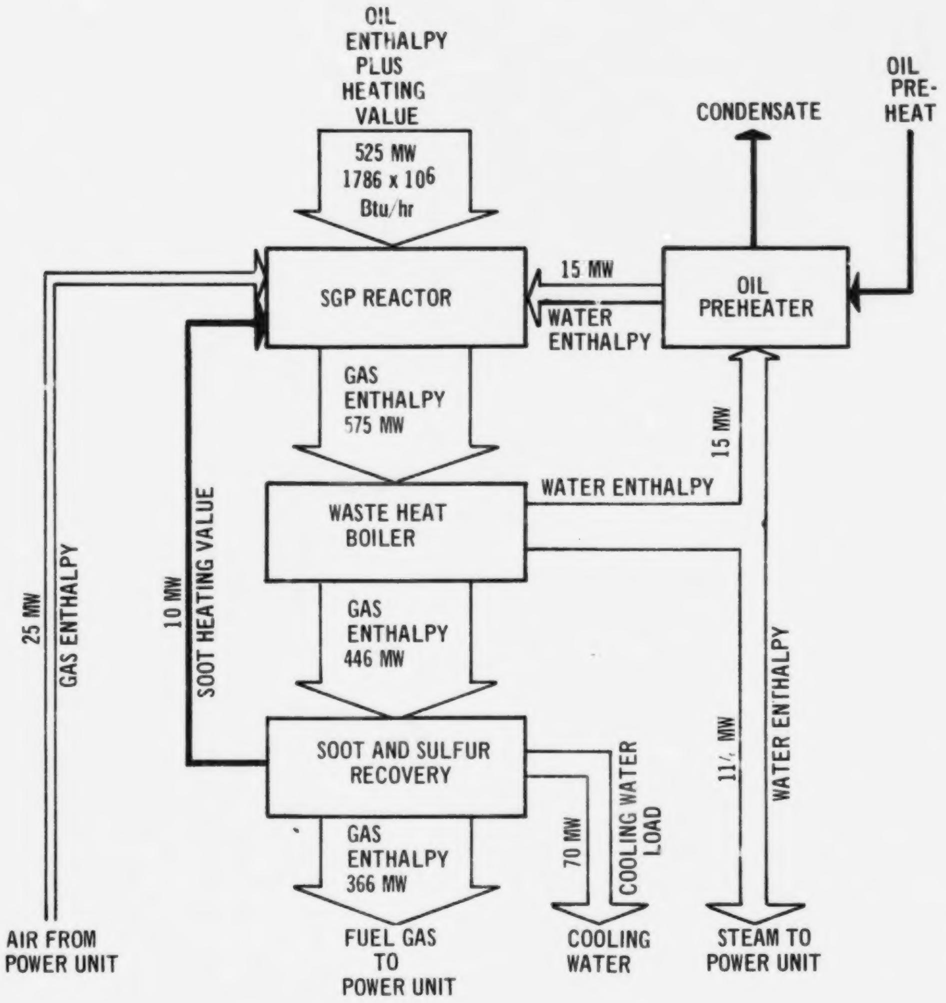


Figure 4A. Power flow through SGP.

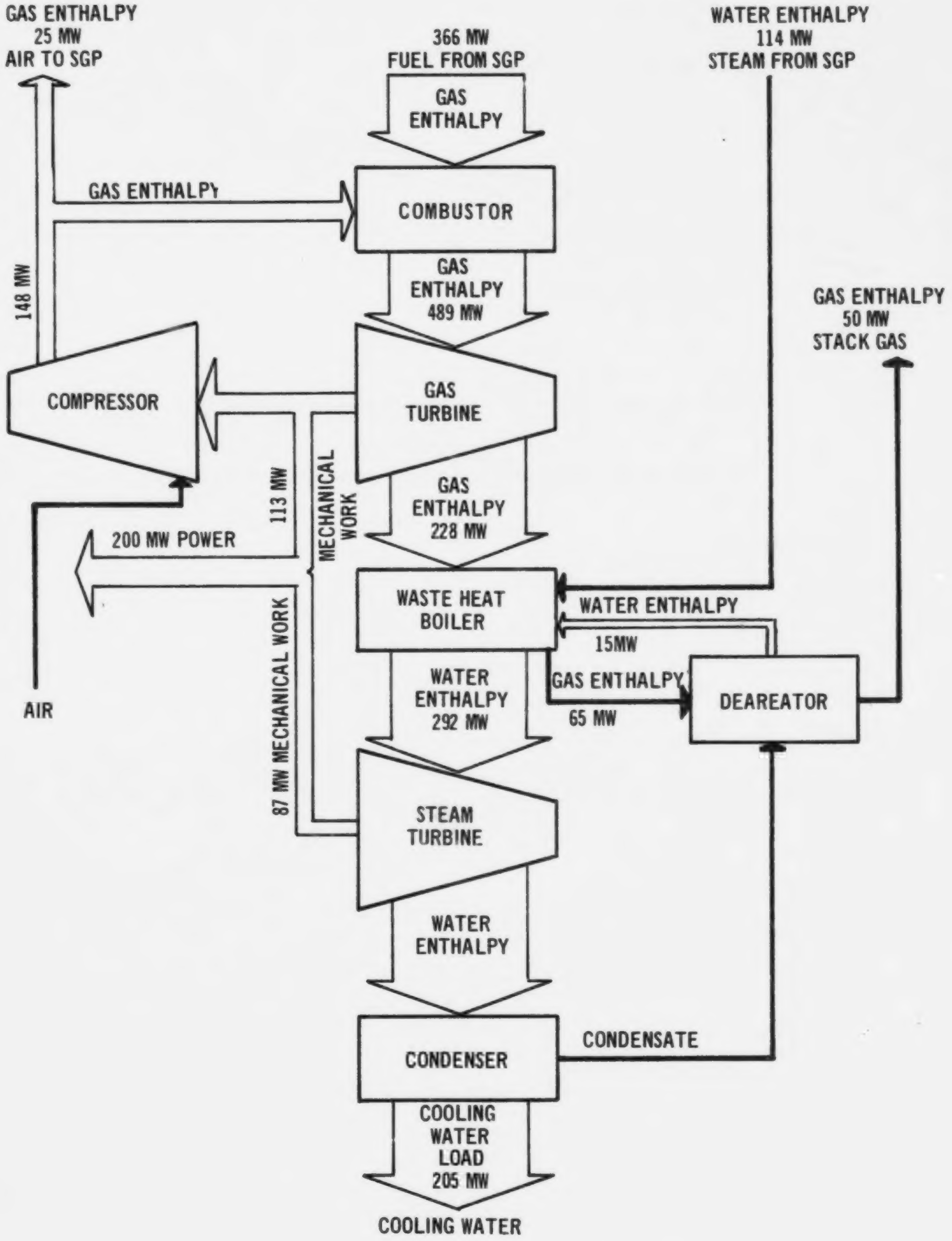


Figure 4B. Power flow through power plant.

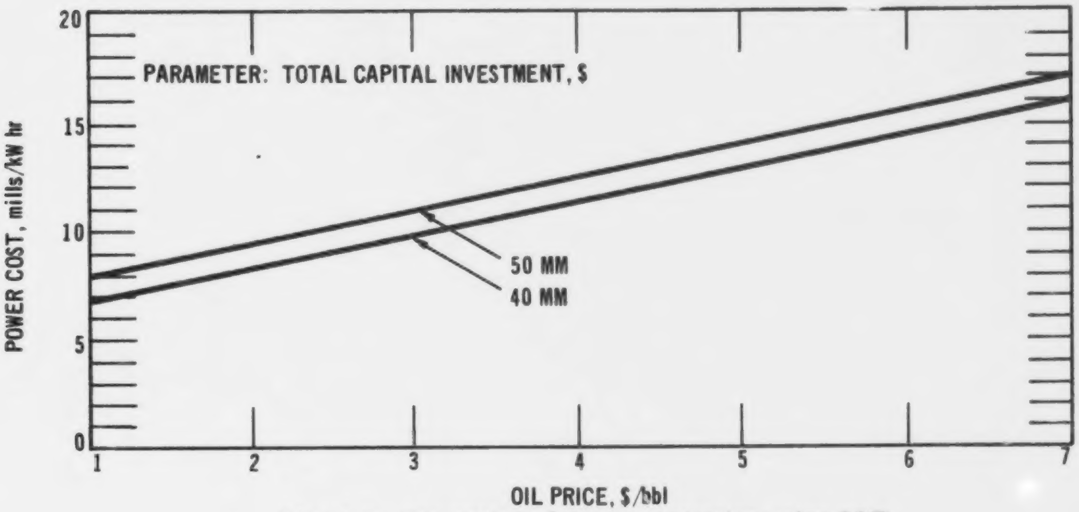


Figure 5. Economics of power generation using SGP.

5. FLUIDIZED-BED OIL GASIFICATION FOR CLEAN POWER GENERATION—ATMOSPHERIC AND PRESSURIZED OPERATION

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ABSTRACT

This paper evaluates high sulfur residual oil gasification for the purpose of clean power generation. It also considers both atmospheric pressure operation with conventional boilers and pressurized operation utilizing combines operation with conventional boilers and pressurized operation utilizing combined steam and gas turbine cycles.

The outlook and status of atmospheric pressure fluidized-bed oil gasification is reviewed. The process has been studied by Esso (England) on a 1-MW pilot plant scale and has been shown to be an effective pollution control device. Preliminary cost estimates for retrofit systems on conventional plants indicate a potential energy cost reduction of 30 to 50 percent over wet scrubbing or low sulfur fuel alternatives. A 30 to 100-MW demonstration plant is scheduled.

A pressurized fluid-bed oil gasification process has been proposed, and preliminary assessment is being carried out in which plant performance, capital costs, and energy costs are examined. The projected ability of the fluid-bed process to meet both pollution regulations and gas turbine requirements is based on Esso (England) atmospheric pressure data. Capital and energy costs for the pressurized fluid-bed combined-cycle (PACE) plant process are compared with a conventional oil-fired plant with wet scrubbing, pressurized fluid-bed combustion of oil, PACE plant operation with No. 2 distillate fuel oil, and conventional pressurized oil gasification processes developed by Shell and Texaco. The fluid-bed process has the potential to reduce energy costs > 20 percent below a conventional plant with wet scrubbing and > 15 percent below a PACE plant using distillate fuel oil.

INTRODUCTION

Electric utility demand for residual oil is projected to increase by around 250 percent by 1980. Low sulfur residual to meet Federal and local regulations will not meet the demand without an intense effort on desulfurization.

Gas producers for making low Btu gas from oil are becoming more attractive to the electric power industry as the availability and cost of natural gas and clean fuel oils decreases and because the technology for removing sulfur dioxide from stack gases is proving to be expensive. A gasification system must provide a clean gas, reliability, utilization of a wide range of fuel, competitive energy cost, and high efficiency.

Fluidized-bed oil gasification can be applied to power generation to produce a clean, low-Btu fuel gas—200 to 500 Btu/scf. In a fluidized-bed gasifier, oil is added to a fluidized-bed of limestone or dolomite with sufficient air—15 to 25 percent of stoichiometric—to maintain the bed at ~ 1600°F and react with the oil to produce a fuel gas. The limestone or dolomite removes sulfur from the fuel gas during the gasification process.

Gasification of oil can be carried out at either atmospheric or elevated presure. Operating at atmospheric pressure, a fluidized-bed gasifier provides clean fuel to a combined cycle gas and steam turbine power plant. The potential for high efficiency and low capital cost of such a plant makes this system attractive.

It has been demonstrated that oil can be gasified and sulfur removed from the resulting fuel gases in a fluidized bed. The design and evaluation of a fluidized-bed oil gasifier operating at atmospheric pressure have been completed. A 30 to 100-MW demonstration plant is scheduled. A pressurized fluidized-bed oil gasification system has been designed. The economics and performance of a pressurized oil gasification combined cycle power plant have been evaluated.

ATMOSPHERIC PRESSURE FLUID-BED OIL GASIFICATION

Westinghouse is evaluating the gasification/desulfurization of residual oil at atmospheric pressure under contract to the Office of Research and Monitoring* (ORM) of the Environmental Protection Agency. The concept is being studied for the purpose of producing, on-site, a low-sulfur fuel gas suitable for power plant utilization in a conventional boiler. Esso Petroleum Company has provided experimental information on the oil gasification/desulfurization process based on small scale batch fluidized units and a 750-kW continuous unit. Westinghouse has carried out preliminary conceptual design studies to evaluate the commercial process, and is presently attempting to locate a utility partner with which to carry out a demonstration plant operation.

Gasification/Desulfurization Concepts

Two possible modes for gasification/desulfurization operation are the regenerative mode and the once-through mode. Figure 1 illustrates the major process streams and identifies the basic elements of the two operational modes.

The elements of the regenerative operation are the gasifier vessel and the regenerator vessel. The gasifier is an air-fluidized bed of lime operated at 1600°F with sub-stoichiometric air, ~20 percent of stoichiometric. The oil is injected into the gasifier vessel where it cracks and is partially combusted to form a hot, low-sulfur fuel gas. Hydrogen sulfide is produced during the gasification which reacts with the lime to produce a sulfided lime.

$$H_2S + CaO \rightarrow CaS + H_2O \qquad (1)$$

The fuel gas is transported to the boiler burners where combustion is completed and the sulfided lime is sent to the regenerator. The regenerator is an air-fluidized vessel operated with a slight excess of air at 1900°F. Regeneration takes place by reaction of oxygen with the utilized lime to give an SO_2 rich stream (of about 10 mole percent SO_2) and a regenerated lime having a slightly decreased activity compared to that of fresh lime.

$$CaS + \frac{3}{2} O_2 \rightarrow CaO + SO_2 \qquad (2)$$

The SO_2 stream is transported to a sulfur recovery system and the regenerated lime is returned to the gasifier along with a stoichiometric amount of fresh make-up limestone.

The elements of the once-through operation shown in Figure 1 consist of a gasifier vessel and a sulfate generator, or predisposal vessel. The operation of the gasifier for once-through limestone operation is the same as for the regenerative operation. The sulfate generator operates similarly to the regenerator, but at a lower temperature (~1500°F) so that the sulfided lime from the gasifier is converted to calcium sulfate rather than calcium oxide. The dry calcium sulfate may be disposed of while the gas stream from the sulfate generator is sent to the gasifier. A limestone addition rate of 3 to 4 times that used in the regenerative operation is necessary in the once-through operation to achieve similar sulfur removal of 90 to 95 percent.

Experimental Work

Under contract to the Office of Research and Monitoring, Esso (England) is carrying out laboratory tests on atmospheric-pressure batch fluidized-bed equipment to investigate lime sulfur absorption and lime regeneration operating variables.[1,2] The results of the Esso Petroleum Company experimental program have identified the critical phenomena associated with atmospheric-pressure gasification/desulfurization, and have established the probable operating conditions and behavior of a commercial system. This evaluation has been based on the results of the small scale batch work. The batch work has been carried out in conjunction with the construction and operation of a 750-kW continuous pilot unit.

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*Previously under the Office of Air Programs.

Design

Energy and material balances provide the basic information with which the feasibility of applying the gasification/desulfurization concept as an add-on to an existing boiler, or as a new plant design feature, has been examined. The feasibility of the retrofit concept has been examined in terms of the availability of space in an existing power plant, the modifications necessary to retrofit an existing boiler, and the performance of a modified boiler. The specifications assumed for the conceptual design are presented in Table 1.

The feasibility of converting an existing boiler to one which utilizes the fuel from a gasification/desulfurization system depends on a number of factors, many of which will differ from one boiler to the next. The space available for a gasification/desulfurization system in an existing power plant, the modification necessary to retrofit an existing boiler, and the performance of a retrofit boiler will depend on the specific gasification/desulfurization system design and choice of operating conditions, the location of the gasification/desulfurization system in the plant, the type of boiler (coal-, oil-, or gas-fired), size of boiler, the turn-down needed, the boiler load factor, and the specific design features of the boiler.

The gasification system may be placed internal to the boiler (directly beneath) or external to the boiler. Preliminary considerations seem to favor the external retrofit design over the internal retrofit design, and the feasibility of retrofitting a coal-fired boiler over the feasibility of retrofitting an oil- or gas-fired boiler. The internal design concept appears to be limited by the available space beneath the boiler and the cost for modifying the boiler. The external design concept should reduce modification costs and should allow greater uniformity and flexibility in system design. Coal-fired boilers have more potential space near the boiler than oil- or gas-fired boilers and may provide some of the needed solids handling and particulate clean-up equipment. Boiler derate should be of little concern with coal-fired boilers because they are designed to operate with slag on the boiler heat transfer surface. Derate may be a concern with oil- and gas-fired boilers due to a reduction in the flame temperature and a

Table 1. SPECIFICATIONS FOR CONCEPTUAL DESIGN

Process specifications	Design variables
Sulfur removal: 90-95% Fuel oil: 3 wt % sulfur; LHV = 17,700 Btu/lb Limestone: 98.6% CaO yield	Boiler size: 600 MW Load factor: 40%, 80% Turn-down: 4/1

Operating variables

	Regenerative system	Once-through system
Gasifier temperature	1600°F	1600°F
Regenerator (sulfate generator) temperature	1900°F	1500°F
Stone make-up rate	1 mole CaO/mole sulfur	3 moles CaO/mole sulfur
Air/fuel ratio	20% of stoichiometric	20% of stoichiometric
Limestone utilization	∼5 wt % sulfur in bed	∼ 19 wt % sulfur in bed
Fluidization velocity	8 ft/sec	8 ft/sec
Minimum fluidization velocity	3 ft/sec	1 ft/sec
Particle sizes (avg)	∼2000 μm	∼ 1000 μm
Gasifier bed depth (static)	2.5-3.5 ft	3.5-4.0 ft

redistribution of the heat release within the boiler which may occur with the fuel.

In contrast to boiler retrofit considerations, the feasibility of incorporating a gasification/desulfurization system into a new boiler design will be limited only by the overall economics of the system and the market potential for new oil-fired boilers. The total space occupied by the gasification/desulfurization system will be a small percentage of the total plant volume. Also, because of the flexibility in boiler design, boiler performance will not be affected by the presence of the gasification/desulfurization system.

Figure 2 shows a plant layout for a 600-MW once-through gasification/desulfurization system. The plant consists of two gasification modules, each utilizing air from one of the two power plant forced draft fans present in the existing boiler.

Evaluation

The design study, coupled with the experimental work of Esso Petroleum Company, points out the technical and economic feasibility of oil gasification/desulfurization as a retrofit SO_2 control system for utility boilers, or as an SO_2 control system to be incorporated into a new boiler design. A market for retrofit and new oil-fired boilers exists.[3]

Preliminary investigations indicate that, overall, the once-through operation may be somewhat more attractive to a utility customer than the regenerative operation. Capital investment is reduced with the once-through operation and, although the limestone feed rate is expected to be three times the rate with regenerative operation, the operating costs for once-through operation may be less than those for the regenerative operation. A complete cost breakdown for once-through and regenerative operation has been presented.[4] Once-through operation has fewer technical problems at this time, and is an overall simpler process than regenerative operation.

A comparison is made in Table 2 between atmospheric pressure oil gasification/desulfurization and the alternative schemes of low-sulfur oil and stack gas cleaning. Capital costs and operating costs are compared for new and retrofit systems. Oil gasification/desulfurization compares favorably with low-sulfur oil and stack gas cleaning, based on prelimimary cost estimates. A reduction of about 40 percent in the capital costs involved in stack gas cleaning is estimated for new and retrofit gasification/desulfurization systems. Operating costs appear to be about the same for once-through stack gas cleaning and regenerative gasification/desulfurization with sulfur recovery. Once-through gasification/desulfurization may reduce the operating costs 30 to 50 percent as compared to stack gas cleaning. The cost estimate indicates that the operating cost with low-sulfur fuel oil will be about 30 to 50 percent greater than the operating cost for gasification/desulfurization. These conclusions are based on the desulfurization of high-sulfur residual oil (3 weight percent sulfur) and may be altered when a lower sulfur oil is considered (1 to 1.5 weight percent sulfur). Environmental factors are also compared in Table 2. The low-sulfur oil is advantageous in that capital costs are limited to possible boiler modifications necessary when changing from gas or coal to low-sulfur oil. On the other hand, operating costs are higher than those for stack gas cleaning or oil gasification/desulfurization. Capital costs are extremely high with stack gas cleaning, especially on the retrofit case, while operating costs are very near those estimated for oil gasification/desulfurization.

Advantages of atmospheric-pressure oil gasification over stack gas wet scrubbers include:

Corrosion and fouling problems minimized in SO_2 removal process and boiler (minimum SO_x and V),

No flue gas reheat required,

Uses crushed limestone — no limestone pulverizing system needed,

Table 2. ASSESSMENT OF FLUIDIZED BED OIL GASIFICATION/DESULFURIZATION

	Low-sulfur oil	Stack gas cleaning	Oil gasification	
			Regenerative operation	Once-through operation
Cost				
Capital, \$/kW				
New	—	25-40	12-15	8-10
Retrofit	a	40-75	22-27	18-22
Fuel adder, ¢/10^6 Btu				
New	25-35	11-14	9.5-16.0	9-10.5
Retrofit	25-35[a]	14-20	11-18	10.5-12.5
Efficiency (thermal)	—	0.95-0.98	0.89-0.96[b]	0.96-0.97[b]
Environmental factors				
SO_2, lb/10^6 Btu	0.35	0.45	0.35	0.35
NO_x, lb NO_2/10^6 Btu	0.40	0.8	0.16	0.16
Particulates, lb/10^6 Btu	0.06	0.05	0.02-0.2[c]	0.02-0.2[c]
Solid waste, ft^3/MW-day	—	25	15	45
Sulfur removal				
Stone	NA		Limestone	Limestone
Ca/S	NA		~ 15[d]	3.0
Make-up Ca/S	NA		1.0	NA

[a] Equipment modifications are required when converting from gas or coal to low sulfur oil.

[b] Overall efficiency is dependent on mode of temperature control.

[c] 0.02 figure based on installing electrostatic precipitator (ESP).
0.2 figure based on installing high efficiency cyclone before burners and no ESP.

[d] Ca/S rate dependent on regenerator temperature control scheme.

Basis: 3% Sulfur, 90% Sulfur Removal, 600 MW Capacity, 8% Load Factor

Simplified disposal — dry solids and no disposal pond,

More compact system,

Reduced structural costs,

Lower auxiliary power requirement,

Reduced energy cost,

Improved NO_x control, and

Potential market for spent CaO.

DEMONSTRATION PLANT PROGRAM

A three phase demonstration plant program has been conceived.

Phase I. Preliminary design and cost estimate of demonstration plant installed on an existing boiler.

Phase II. Detail design and construction of demonstration oil gasification process.

Phase III. Developmental operation of the gasification process and integrated power plant.

A utility (or utilities) is required as a cooperating party to carry out this program. In Phase I the utility would supply technical information on its existing power plant, provide price and availability information on oil fuels which might be utilized in the plant currently and/or in the future, supply information concerning the load requirements placed on the plant, cooperate in the selection of an engineering firm to prepare the preliminary design, assist in selecting various options in the design of the system and evaluate, in cooperation with Westinghouse, Esso (England) and EPA, the effectiveness and economy of the oil gasifier/desulfurizer in power generation and pollution abatement.

An engineering firm will carry out designs in sufficient detail that fixed price bids can be solicited for detailed design and construction of the system. The design effort will be based on experimental data from Esso (England) on their 1-MW continuous pilot plant and the conceptual design and assessment by Westinghouse. EPA will provide general guidance and funding for Phase I.

If the preliminary design confirms the effectiveness and economics of the system, a proposal would be prepared for Phases II and III. In Phase II the utility would work closely with the engineering firm in the design and installation of the gasifier/desulfurizer system and share in the costs of the design and installation.

In Phase III the utility would operate the plant, collect basic data on the operation of the gasifier/desulfurizer system, aid in its interpretation and analysis, and cooperate in the evaluation of the effectiveness, technical and economic, of the process in power generation and pollution abatement.

Presentations have been made to 13 utilities who currently operate or plan to operate oil-fired power plants. Presentations began in March 1972 and have included utilities from the East Coast, West Coast, Florida and the South Central U.S. A West Coast utility is actively interested in the program. Seven utilities are currently evaluating the proposal, and five utilities have indicated they are not interested in participating in the program at this time.

Problem areas for which utilities have expressed concern are:

1. Hot fuel gas piping and valves.
2. Control and emergency conditions.
3. Space requirements.
4. Solid waste disposal.
5. Modifications and time required for modifications to boiler.
6. Availability.

These and similar technical concerns will be evaluated in detail during the preliminary design phase. In addition, the prospect of obtaining funds from EEI (Edison Electric Institute) has been pointed out.

Conclusions

Performance

The concept has been technically demonstrated with a 750-kW development gasification plant.

Sulfur removal up to 95 percent can be achieved.

Nitrogen oxide emissions of 150 ppm appear possible.

Particulate emissions will be higher than conventional oil- or gas-fired systems but can easily be removed to achieve proposed standards.

Further development effort is required in key areas—e.g., calcium sulfate generation for once-through operation, temperature control, sulfur recovery.

Economics for comparable pollution abatement

Capital cost of a retrofit, once-through gasification system may be 50 to 70 percent less than a retrofit wet scrubbing system.

Fuel adder cost for a retrofit, once-through gasification system may be 30 to 50 percent less than wet scrubbing, low-sulfur oil, or desulfurized oil.

Market

Initial market is expected to be small boilers (<600 MW) on the East Coast, in the Southwest where gas may be limited, or on the West Coast. Once-through operation may be favored over operation with sulfur recovery for these plants.

PRESSURIZED FLUID-BED OIL GASIFICATION

Two different processes are being considered for the pressurized gasification of residual oil: (1) a pressurized version of the fluidized bed oil gasification/desulfurization process, being developed by Esso (England), which has been explored only at atmospheric pressure, and (2) the pressurized oil gasification process of the type which has been operated for gas manufacture by Shell[5] and Texaco.[6] The Shell and Texaco processes are not identical, but they are very similar in concept, performance and cost. The process concept is the important factor for this study so the Shell and Texaco processes are not discussed individually. The Shell and Texaco processes generate a low temperature (100-250°F), clean fuel gas having a low heating value of about 120 Btu/scf. Steam generated by waste heat boilers in the gasification process is also provided for the combined cycle plant. The fluidized-bed process generates a hot ($\sim$1600°F), clean fuel gas having a low heating value of 200 to 500 Btu/scf (hot). Preliminary cost and performance estimates for these two process concepts have been developed and are compared with alternative oil-fueled power generation techniques having pollution control.

Process Concepts and Options

Flow diagrams for the Shell and Texaco processes and the pressurized fluid-bed process are shown in Figures 3 and 4, respectively. The Shell and Texaco processes consist of an air-blown oil gasification vessel (partial oxidation reactor) operated at a temperature of about 2500°F with an air/fuel ratio of about 45 percent of stoichiometric. The hot gas is cooled in waste heat boilers, producing saturated steam, prior to purification of the gas. The gas purification process consists of a carbon (soot) recovery step and a sulfur removal section. Recovery soot ($\sim$ 3 weight percent of the fuel oil feed rate) is recycled to the gasifier vessel, and H_2S produced in the sulfur removal section is sent to a sulfur recovery plant to recover elemental sulfur.

The pressurized fluid-bed oil gasification process shown in Figure 4 consists of a fluidized bed gasifier/desulfurizer vessel, a limestone/dolomite regeneration section, and a sulfur recovery section. The phenomena taking place in the pressurized fluid bed gasifier are essentially the same as have been described for the atmospheric pressure fluid bed case. The gasifier is operated at about 1600°F with an air/fuel ratio of about 14-25 percent of stoichiometric. Both limestone and dolomite are considered as sulfur absorbents in the pressurized fluid-bed case. The major process options which have been examined with respect to cost and performance are:

1. The gasifier air/fuel ratio. Air/fuel ratios of 14 percent and 25 percent of stoichiometric giving fuel gas low heating values (hot) of about 500 and 280 Btu/scf, respectively, have been considered. An air/fuel ratio of 14 percent of stoichiometric is assumed to be the minimum air/fuel ratio at which a gasifier temperature of 1600°F can be maintained, while 25 percent of stoichiometric

is assumed to be a conservatively high air/fuel ratio according to the experience gained from the Esso (England) atmospheric pressure operations. The physical feasibility of operating at an air/fuel ratio as low as 14 percent of stoichiometric without excessive carbon deposition in the gasifier must be demonstrated.

2. The limestone/dolomite regeneration method. The regeneration of calcium suflide and the production of a high sulfur gas for sulfur recovery can be achieved by either of two processes:

Regeneration with CO_2 and H_2O to produce H_2S. With this process the reaction

$$CaS + H_2O + CO_2 \rightleftharpoons CaCO_3 + H_2S \qquad (3)$$

is utilized to produce calcium carbonate and H_2S-rich gas. The reaction is favored by high pressure and would be carried out at a temperature of about 1100°F, with H_2S concentrations of about 9 percent by volume being projected. Two sources of CO_2 for the regeneration scheme have been considered--CO_2 provided by scrubbing flue gas from the combined cycle plant, and CO_2 provided by scrubbing a gas stream produced by combustion of a portion of the fuel gas.

Regeneration with air to produce SO_2. The reaction of oxygen with calcium sulfide,

$$CaS + 3/2\,O_2 \rightleftharpoons CaO + SO_2 \qquad (4)$$

is favored by low pressure and high temperature ($\sim$2100°F). Although this regeneration scheme would yield an SO_2 concentration of only about 2 percent by volume at pressures of 10-15 atm, it is considered because of its apparent simplicity.

If a high stone make-up rate is required for the regenerative processes and if high stone utilization can be achieved in the gasifier, a once-through system may be attractive. Once-through operation would require conversion of calcium sulfide to calcium sulfate before disposal of the stone. The reaction

$$CaS + 2\,O_2 \rightleftharpoons CaSO_4 \qquad (5)$$

could be applied for this purpose, and would be carried out at 1400-1700°F. Limestone utilization in a once-through process is expected to be 35 percent or higher.

Other process options examined are the pressure drop required across the gas turbine combustor and the option of cooling the gas produced by fluidized-bed oil gasification before it is combusted in the combined cycle plant.

Process Specification and Design Basis

Table 3 lists the factors specified for the conceptual design of the pressurized fluid-bed oil gasification process. The specifications for the plant capacity, capacity factors, and the turndown ratio are also assumed for the Shell and Texaco processes. The combined cycle power generating plant is a Westinghouse PACE Plant consisting of two Westinghouse-501B gas turbines and a single 28.5-in. steam turbine. The pressure of the fuel gas to the gas turbine is 215 psia. Designs and energy costs are based on operation using limestone for desulfurization. Sulfur removal of 90 to 95 percent from a 3 weight percent sulfur residual oil

Table 3. SPECIFICATIONS FOR FLUID-BED OPERATION

Plant electrical capacity	250 MW (PACE Plant)
Capacity factor	70%
Turndown ratio	4/1
Number of gasifier modules	2
Modes of operation	Regenerative or once-through
Gasifier pressure	$\sim$15 atm
Pressure of gas turbine	11 atm (165 psia to turbine)
Sulfur removal	90-95%
Residual oil	3 wt % sulfur
Gasifier temperature	1600°F
Regenerator temperature	1100°F[a]
Sulfate generator temperature	1700°F with once-through option
Air/fuel ratios	14 (minimum) & 25% of stoichiometric
Lime particle diameters	500-2000 μm average diameter
Regenerative lime utilization	10%
Once-through lime utilization	35%
Limestone make-up rate	1 mole CaO/mole sulfur fed
Gasifier temperature control	Stack gas recycle, steam or water injection
Regenerator temperature control	Lime circulation rate; water injection
Sulfate generator temperature control	Excess air circulation ($\sim$ 200% excess)
Plant heat rate	9000 Btu/kWhr (assumed for purposes of material balances)

[a] With Co_2/H_2O regeneration; 2100°F with air regeneration.

is specified for the fluid-bed process, with specifications for vessel temperatures, limestone utilizations, and limestone make-up rate based both on thermodynamic information and the atmospheric pressure data of Esso (England). Control of the vessel temperatures is assumed to be easily carried out by any of the methods suggested in the table, based on atmospheric information. A plant heat rate of 9000 Btu/kWhr was assumed for the purpose of finding the approximate fuel consumption of a PACE Plant.

Shell and Texaco both supplied energy and material balance information for their processes along with capital investment estimates. The Shell and Texaco processes had not necessarily been modified to provide the optimum power generation performance, but were based mainly on gas manufacturing experience.

Material and energy balance information for the pressurized fluid-bed oil gasification process was based on Esso (England) atmospheric pressure data. Performance and vessel sizes were modified for effects of pressure. Regeneration system designs were based largely on thermodynamic behavior. The general behavior of the fluidized-bed gasifier for sulfur removal, vanadium removal, and carbon deposition is assumed to be independent of pressure once the gasifier bed diameter and bed depth has been scaled for the affect of pressure.

Material and Energy Balances

Figures 5 and 6 show simple block flow diagrams for the Shell and Texaco processes and the pressurized fluid-bed oil gasification process, respectively. Block diagrams of this type have been utilized to analyze the performance of the gasification processes and the performance of the complete power plant.

Inputs to the Shell and Texaco processes, Figure 5, are shown to be residual oil, air, water, booster compressor power, and auxiliary power for pumping oil, boiler water, cooling water, scrubber recycle, etc. Output is cold fuel gas, steam, sulfur, and energy losses. Energy losses for the Shell and Texaco processes arise from heat losses, cooling water losses, and sensible heat of flue gas from gas purification and sulfur recovery sections. The thermal efficiency of the Shell and Texaco processes as presently conceived is less than 80 percent; though improvements in this performance factor may be made by simple process alterations. Shell has recently indicated 87 percent thermal efficiency for their process.

Inputs to the fluidized-bed process (Figure 6) are residual oil, air, water and steam, limestone (or dolomite), booster compressor power, and auxiliary power for pumping oil, water, solids circulation, and gas compression in the regeneration section of the gasification system. Output is hot fuel gas for the combined cycle plant, sulfur, and energy losses in the form of sensible heat of the spent limestone, heat losses, carbon deposition losses, and sensible heat of flue gas from the regeneration and sulfur recovery sections. The thermal efficiency of the fluidized-bed gasification system will depend slightly on the air/fuel ratio and the regeneration method used, but will be about 90 to 95 percent for all the options considered.

Fuel compositions and heat values are shown in Table 4 for the two oil gasification processes. Shell provided expected product

Table 4. GASIFICATION PRODUCT COMPOSITIONS

	Shell process, Vol %	Fluidized bed process, Vol %	
		14 A/F	25 A/F
N_2	60.79	50.74	47.74
H_2	14.57	0.82	2.64
CO	22.92	13.40	7.97
CO_2	1.37	6.70	7.97
H_2O	0.00	0.00	20.56
CH_4	0.35	9.43	6.55
C_2H_4	0.00	18.86	6.55
H_2S	0.00	0.05	0.02
Low heating value (hot), Btu/scf	117	∼500	∼280

compositions for a dry gas, while the product gas compositions for the fluidized-bed process at air/fuel ratios of 14 and 25 percent of stoichiometric have been estimated from atmospheric pressure data. Projected plant performance has been based on these product compositions.

Capital Investment Evaluation for the Pressurized Fluid-Bed Process

Figure 7 is a flow diagram for the pressurized fluid-bed oil gasification process with limestone regeneration be reaction with CO_2 and H_2O to form $CaCO_3$ and H_2S. The source of CO_2 is flue gas from the combined cycle plant. The alternative CO_2 source, combustion of a portion of fuel gas, has been eliminated based on the comparative economics of the two options. The equipment shown in Figure 7 consists of fuel oil and limestone handling equipment, air booster compressor, gasifier vessels (2 modules) with multi-stage particle collection, and a CO_2/H_2O regeneration system.

The process has been separated into four component systems—the gasification system, the CO_2/H_2O regeneration system, the booster compressor, and the Claus plant. These four component costs are shown in Figure 8, in units of $\$/10^6$ Btu-hr of oil feed (HHV), as a function of the pressure at the gasifier product gas outlet, with the air/fuel ratio as a parameter. Two cases of pressure drops required in the gas turbine combustor and the fuel distribution equipment are considered in the figure—8 psi ΔP and 53 psi ΔP. The pressure drop across the gasifier and particle control equipment is assumed to be 9 percent of the gasifier pressure.

Figure 8 leads to the following conclusions:

1. Reducing the air/fuel ratio from 25 to 14 percent of stoichiometric reduces the capital investment by about 10 percent in units of $\$/10^6$ Btu-hr.
2. For the case of a 10 atm gas turbine, increasing the combustor pressure drop from 8 psi to 53 psi reduces the process cost by about 10 percent because of the highly pressure sensitive nature of the CO_2/H_2O regeneration scheme.
3. A once-through scheme would reduce the cost of the process by more than 40 percent, neglecting the slight cost increase due to a sulfate generating vessel.
4. The cost of the fluidized-bed process with an air-blown regenerator would be somewhere between the cost for the CO_2/H_2O regeneration system and the once-through gasification system.

The true capital investment for pressurized fluid-bed oil gasification will depend on the plant performance as a function of the design options--air/fuel ratio, combustor pressure drop, and regeneration method.

Performance

Cycle studies based on estimated material and energy balances and approximate gas compositions (Table 4) have led to estimates of the plant heat rate, the plant power, capital investment, and energy costs, using pressurized oil gasification with combined cycle power generation (PACE Plant). The following conclusions concerning the combustor pressure drop have been deduced:

1. With the pressurized fluid-bed oil gasification process an increase in the required combustor pressure drop of 10 psi increases the plant heat rate by only 10 Btu/kWhr with operation at an air/fuel ratio of 14 percent of stoichiometric, and 30 Btu/kWhr percent.
2. With the pressurized fluid-bed oil gasification process an increase in the required combustor pressure drop of 10 psi reduces the plant power by about 0.5 MW with operation at 14 percent of stoichiometric, and about 1.0 MW at an air/fuel ratio of 25 percent of stoichiometric.
3. With the Shell and Texaco processes, increasing the required combustor pressure drop 10 psi increases the plant

heat rate by about 70 Btu/kWhr, and decreases the plant power by about 3 MW.

From these points it is concluded that the combustor pressure drop is a fairly insensitive parameter with respect to plant performance and should be determined by the requirements for fuel distribution and control.

Table 5 summarizes the performance, capital cost, and energy cost of the Shell and Texaco processes and the fluidized-bed process with a combustor pressure drop of 50 psi for the two air/fuel ratios, and for regenerative operation by CO_2/H_2O, and for once-through operation. The figures shown in the table refer to the case in which the 1600°F fuel gas from the fluid-bed process is not cooled before combustion. Table 5 indicates that the plant heat rate with the fluid-bed process increases with increasing air/fuel ratio, while the heat rate is comparable for the regenerative and once-through operations. The plant heat rate is less than 10,000 Btu/kWhr even for the high air/fuel ratio case of 25 percent of stoichiometric. The Shell and Texaco processes yield a plant heat rate of about 13,000 because of the low thermal efficiency, the high booster compressor power requirements, and the relatively high ratio of power produced by the steam turbine to the power generated by the gas turbine. A plant heat rate of 11,000 Btu/kWhr is estimated for 87 percent thermal efficiency. Plant power is comparable for all of the cases shown, with the Shell and Texaco processes yielding the highest power. Capital investment is based on PACE Plant cost information and the cost estimates shown in Figure 8. The cost basis is listed in Table 5. Capital cost is reduced for the fluid-bed process with once-through operation due to the great expense involved in limestone regeneration by the CO_2/H_2O method. The change in plant capital cost in going from 14 percent air/fuel ratio to an

Table 5. PROCESS PERFORMANCE

	Fluid bed process CO_2/H_2O limestone regeneration 14% A/F	Fluid bed process CO_2/H_2O limestone regeneration 25% A/F	Fluid-bed process once-through operation 14% A/F	Fluid-bed process once-through operation 25% A/F	Shell and Texaco thermal efficiency 75%	Shell and Texaco thermal efficiency 87%
Plant heat rate (HHV), Btu/kWhr	9,007	9,828	8,906	9,716	13,000	11,000
Plant power, MW	266.0	276.0	269.0	279.0	282.4	282.4
Plant capital cost, $/kW	179.5	182.1	158.8	159.5	246.7	246.7
Total energy cost, mills/kWhr	9.43	9.93	8.96	9.42	13.34	12.44
Break-even distillate cost, ¢/10^6 Btu	65.2	71.1	59.7	65.1	111.1	98.7

Conditions Assumed:

1. 50 psi combustor pressure drop.
2. 1600°F fuel gas temperature from fluid-bed process with no cooling.
3. Fixed charges at 15%.
4. Residual oil at 45¢/10^6 Btu.
5. Limestone at $6/ton.
6. 5% contingency; 4% escalation; 8% interest during construction; 2 years construction time; 2% A&E; 70% capacity factor.
7. No credit for sulfur recovered.

air/fuel ratio of 25 percent of stoichiometric is slight. The plant capital investment for the Shell and Texaco processes are based on the estimated gasification system capital cost of about $100/kW provided by both Shell and Texaco.

Energy cost assumptions are listed in Table 5. The 14 percent air/fuel ratio operation of the fluid-bed process is more economical than the 25 percent air/fuel ratio, while the energy cost for once-through operation is also less than regenerative operation. The relationship between energy cost of regenerative and once-through operation is dependent on the cost of limestone disposal, but even if the limestone cost should double to $12/ton the once-through and regenerative system energy costs will be about equal.

Break-even costs for No. 2 distillate fuel oil is given in ¢/10^6 Btu and represent the price at which No. 2 distillate must be available for the PACE Plant to operate at the same energy cost as with residual oil gasification. With No. 2 distillate prices ranging from 85-95¢/10^6 Btu, the fluid-bed process is expected to be more economical than PACE Plant operation with No. 2 distillate. The Shell and Texaco processes, as they are now conceived, do not appear competitive with operation with distillate fuel due to the large capital investment and inefficiency of the gasification system. The possibility of altering the Shell and Texaco processes to make them more compatible with power generation requirements does exist.

The performance of the fluid-bed oil gasification process has been examined with respect to three methods of cooling the 1600°F product gas: (1) cooling the gas and discarding the sensible heat, (2) cooling the gas and recovery of sensible energy at an efficiency of 25 percent, and (3) cooling the fuel gas by direct cooling with injection of 77°F water. The following general conclusions have been derived from this examination:

1. For both regenerative and once-through operations with the fluid-bed process, cooling the 1600°F fuel gas to 400°F and discarding the energy will increase the plant heat rate by about 1600 Btu/kWhr, increase the energy cost by about 0.9 mill/kWhr, and yield nearly the same plant power, with an air/fuel ratio of 25 percent of stoichiometric. The corresponding increases with an air/fuel ratio of 14 percent are about 800 Btu/kWhr and 0.45 mill/kWhr.
2. For both regenerative and once-through operations with the fluid-bed process, injection of water to cool the 1600°F fuel gas to 200°F increases the plant heat rate by about 3,500 Btu/kWhr, increases the plant power by about 140 MW, and increases the energy cost by about 1.0 mill/kWhr, and an air/fuel ratio of 25 percent. The corresponding increases with a 14 percent air/fuel ratio are 1500 Btu/kWhr, 50 MW, and 0.49 mill/kWhr.
3. For both regenerative and once-through operations with the fluid-bed process, cooling the 1600°F fuel gas to 400°F with a waste heat boiler and recovering the energy with an efficiency of 25 percent will increase the plant heat rate by about 500 Btu/kWhr, and increase the plant power by about 40 MW, with a 25 percent air/fuel ratio. The corresponding increases with a 14 percent air/fuel ratio are 300 Btu/kWhr and 15 MW. The energy cost increase is slight if a reasonable cost is assumed for a conventional waste heat boiler system, and if it is assumed that cooling of the hot fuel gas is feasible at air/fuel ratios as low as 25 percent of stoichiometric--about 0.10 mill/kWhr at 14 percent air/fuel ratio and 0.16 mill/kWhr at 25 percent air/fuel ratio. In reality the hot fuel gas cannot be cooled in a conventional waste heat boiler system due to the presence of tars (~5-10 weight percent) which will condense on all cooling surfaces. Cooling of the gas will require either operation at much higher air/fuel ratios (~40 percent of stoichiometric), or quenching and scrubbing of the hot gas to remove tars. The first option would increase the gasifier

capital about $10/kW, not including the cost of a waste heat boiler system and soot removal equipment. The second option would result in inefficient operation with high energy losses, as with cooling of the gas by water injection. Both options seem unattractive for power generation, though further analysis is required to develop quantitative conclusions.

Comparisons With Alternative Power Generation Systems

Tables 6 and 7 compare capital investments and energy costs of pressurized fluid-bed oil gasification with alternative oil-fueled, pollution controlled, power generation systems. Capital and energy costs for a conventional oil-fuel power plant utilizing limestone-wet scrubbing for pollution control, a pressurized fluid-bed combustion plant fueled by oil, and a PACE Plant fueled with No. 2 distillate, are compared with capital and energy costs for a PACE Plant with pressurized fluid-bed oil gasification. The fluid-bed process is carried out at a 25 percent air/fuel ratio with limestone regeneration by the CO_2/H_2O method, representing the highest capital cost and lowest efficiency of the fluid-bed cases considered. Due to the low cost of the PACE Plant package, the PACE Plant with fluid-bed oil gasification is about 140$/kW cheaper than a conventional power plant and 60$/kW cheaper than a regenerative pressurized fluid bed combustion plant fueled with oil. Energy cost of the PACE Plant with fluid-bed oil gasification, assuming 45¢/10^6 Btu residual fuel and 90¢/10^6 Btu distillate fuel, is more than 3 mills/kWhr less then the energy cost of the conventional power plant, about 1.3 mills/kWhr less than pressurized fluid-bed combustion, and about 1.7 mills/kWhr less than a PACE Plant fueled with No. 2 distillate.

Conclusions

1. The pressurized fluid-bed oil gasification process is competitive with alternative oil fueled, pollution controlled, power generation processes. Energy cost reductions of >20 percent are projected. This conclusion holds over the range of factors explored — air/fuel ratios, combustor pressure drops, and limestone regeneration methods.

Table 6. CAPITAL INVESTMENT COMPARISONS

	Conventional oil-fired power plant with scrubber	Pressurized fluid bed oil composition[a]	PACE plant with no. 2 distillate fuel	PACE plant with residual oil gasification[b]
Total capital cost, $/kW[c]	323.28	243.67	137.80	182.1
Assumptions				
Construction time, years	4.5	3.5	2.0	2.0
Sulfur removal equipment, $/kW	50.0	12.68	---	44.3
Plant capacity, MW	635	635	269	269

[a] Operated with limestone regeneration.

[b] CO_2/H_2O regeneration of limestone and air/fuel ratio of 25% of stoichiometric--no cooling.

[c] 5% contingency; 4% escalation per year; 8% interest during construction; 2% A&E; 70% capacity factor.

Table 7. ENERGY COST COMPARISONS
(mills/kWhr)

	Conventional oil-fired power plant with scrubber	Pressurized fluid bed oil combustion	PACE plant with no. 2 distillate fuel	PACE plant with residual oil gasification
Fixed charges	7.91	5.96	3.37	4.45
Fuel	4.11	4.35	7.80	4.42
Limestone	0.12	0.13	---	0.13
Operating and maintenance	0.91	0.82	0.52	0.93
Total	13.05	11.26	11.69	9.93

Assumptions

1. Fixed charges at 15%; 70% capacity factor.
2. 3 wt% sulfur residual oil at 45¢/10^6 Btu.
3. Limestone at $6/ton.
4. No.2 distillate at 90¢/10^6 Btu.
5. No credit for sulfur recovered.

2. Cooling of the 1600°F fuel gas produced by pressurized fluid-bed oil gasification with no recovery of the energy or by water injection is unattractive from the standpoint of capital and energy cost. Cooling to 400°F with 25 percent recovery of the energy may be uneconomical and further analysis is required.

3. An experimental study of pressurized fluid-bed oil gasification should be carried out to determine the process behavior in critical areas and to obtain a better understanding of the economic advantages to be gained from the process.

4. The process concept utilized by Shell and Texaco, though not as efficient or economical as the fluid-bed process concept, should be considered for combined cycle power generation because it depends largely on existing technology. Improvement of the process performance may be possible.

ACKNOWLEDGMENTS

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REFERENCES

1. Moss, G. The Desulfurizing Combustion of Fuel Oil in Fluidized Beds of Lime Particles. (Presented at First International Conference on Fluid-Bed Combustion. Hueston Woods. November 1968.)

2. Craig, J. W. T., G. L. Johnes, G. Moss, and J. H. Taylor. Study of Chemically Active Fluid Bed Gasifier for Reduction of Sulfur Oxide Emissions. Interim Report. Esso Research Centre, Abingdon, Berkshire, England. Prepared for the Air Pollution

Control Office, Environmental Protection Agency, Research Triangle Park, N. C. under Contract Number CPA 70-46. August 1970.

3. Hauser, J. G. Optimum Uses of Energy Sources. (Presented at Spring Conference, Southeastern Electric Exchange. New Orleans. April 1971.)

4. Newby, R. A., D. L. Keairns, and D. H. Archer. Assessment of Fluidized Bed Oil Gasification for Power Generation. (Presented at the 65th Annual Meeting of the Air Pollution Control Association. Miami Beach. June 1972.)

5. Information provided during discussions with Shell, May 3, 1972.

6. Information provided during discussion with Texaco, May 24, 1972.

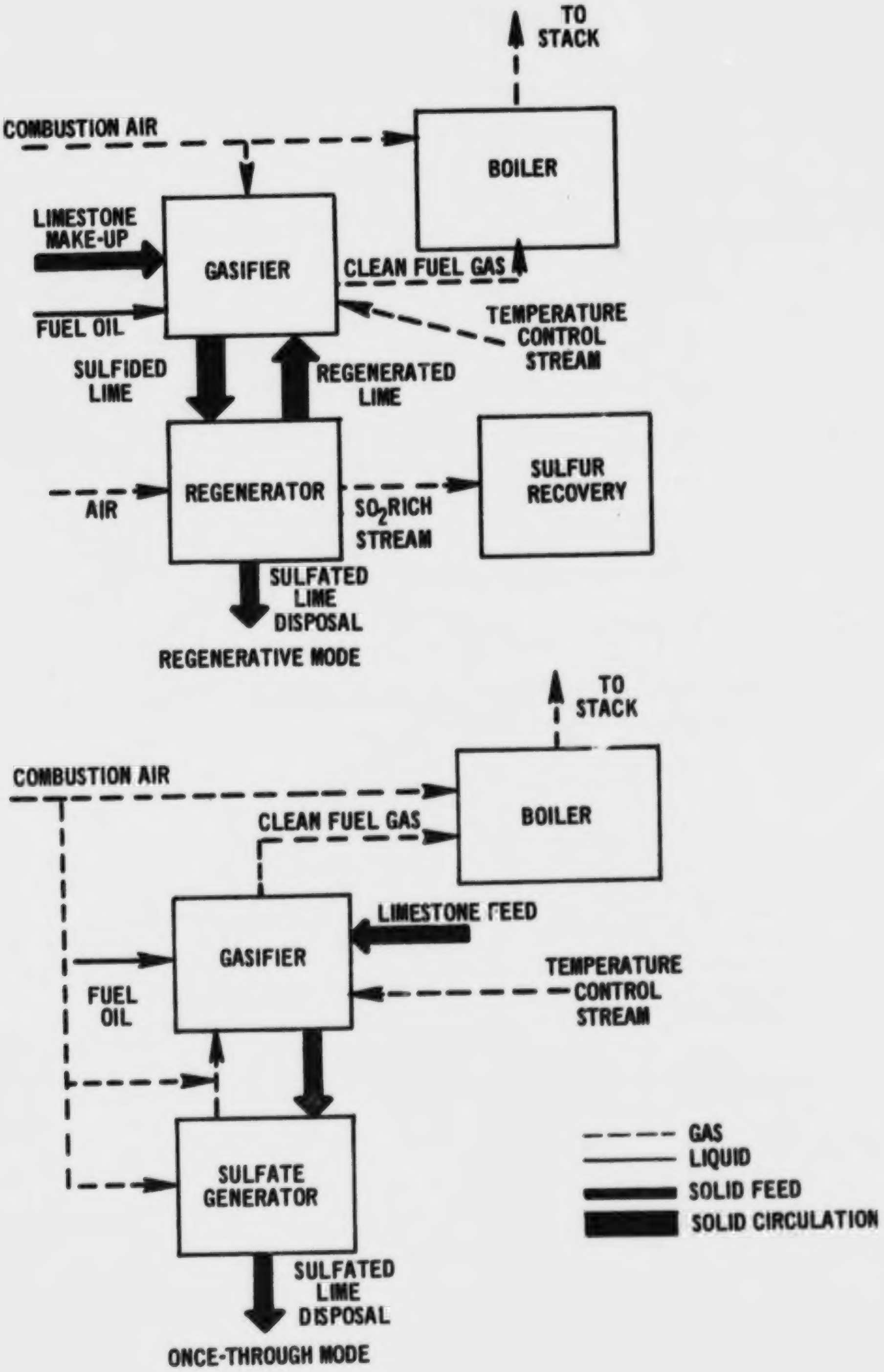

Figure 1. Modes of operation.

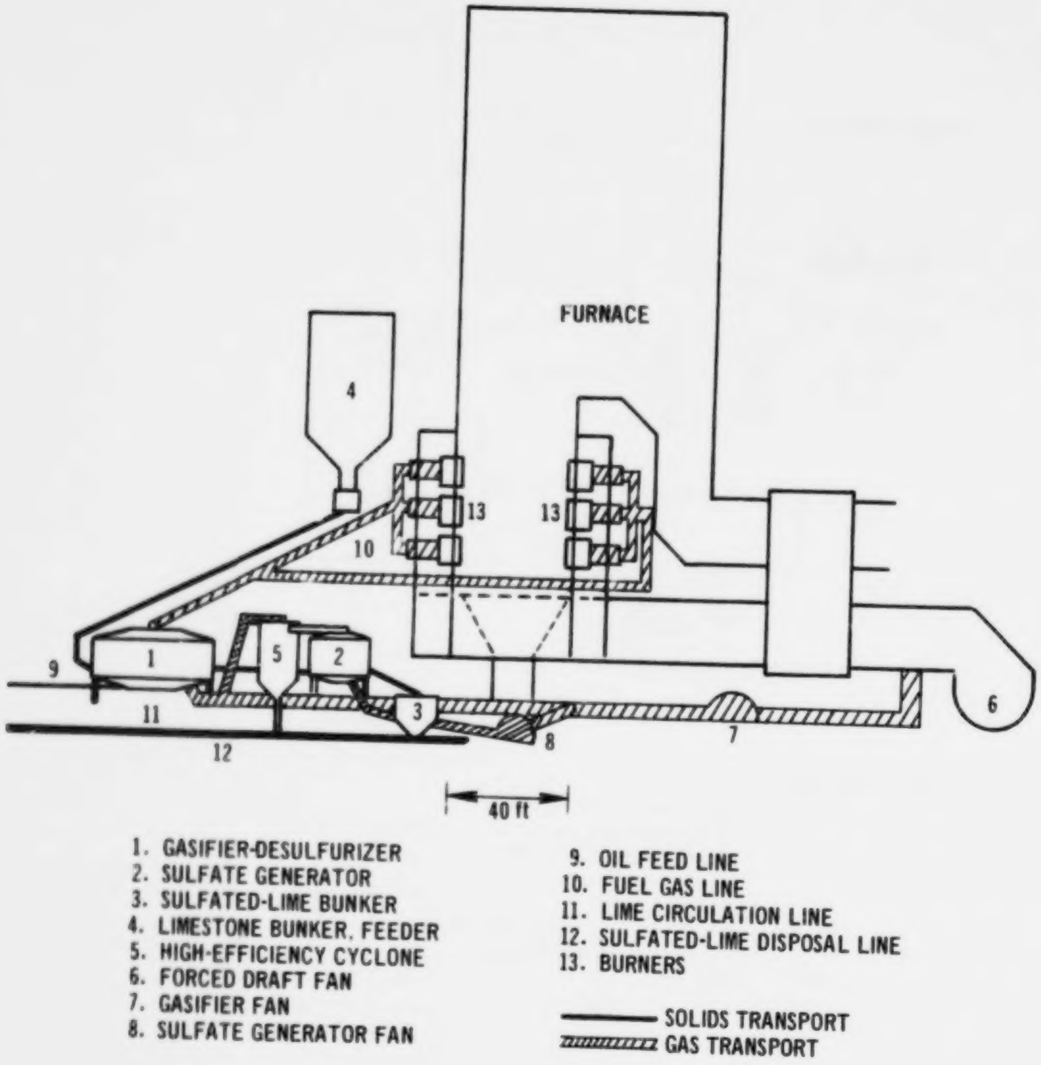

Figure 2. Retrofit of 600-MW boiler, external once-through design.

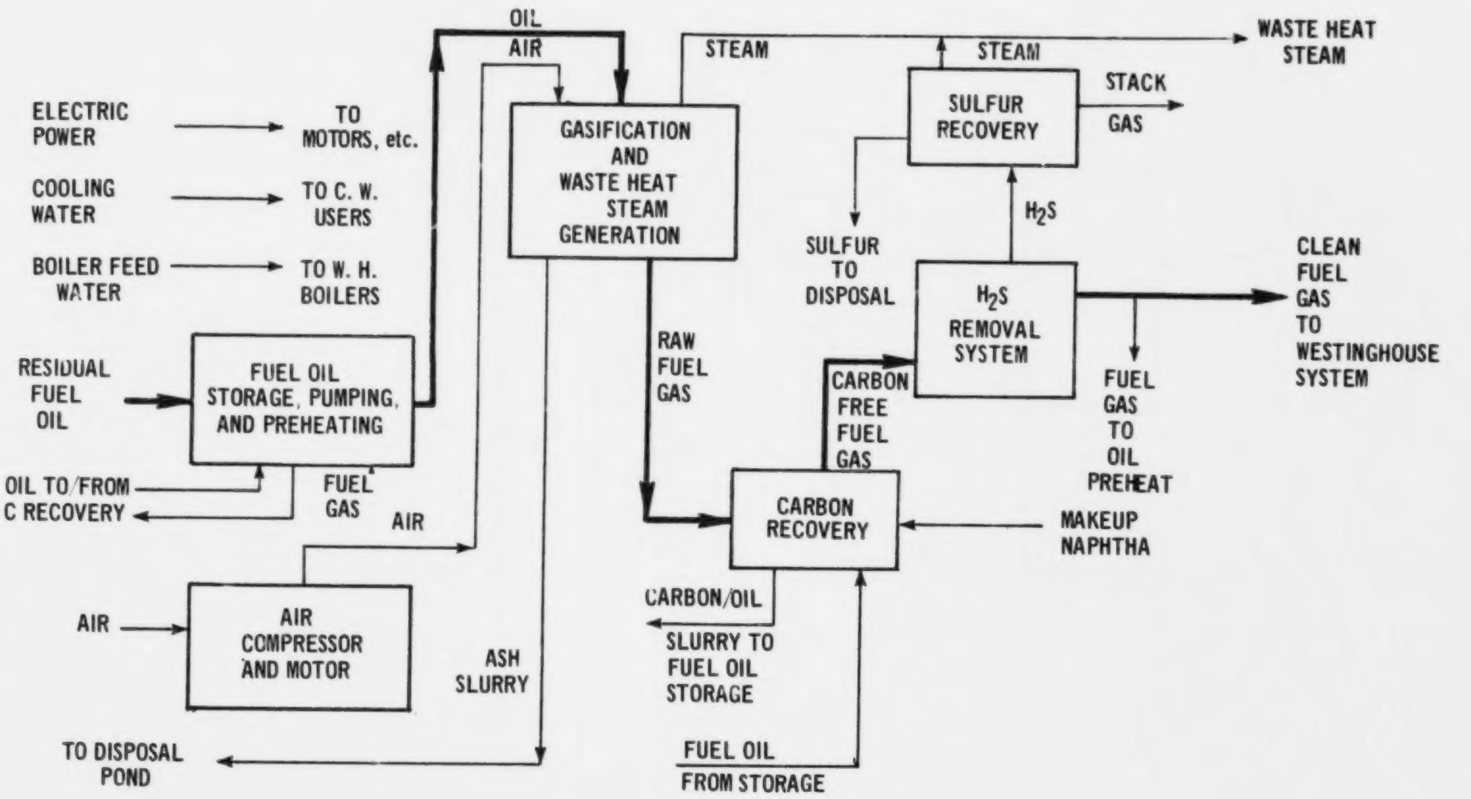

Figure 3. Pressurized oil gasification for gas turbine fuel.

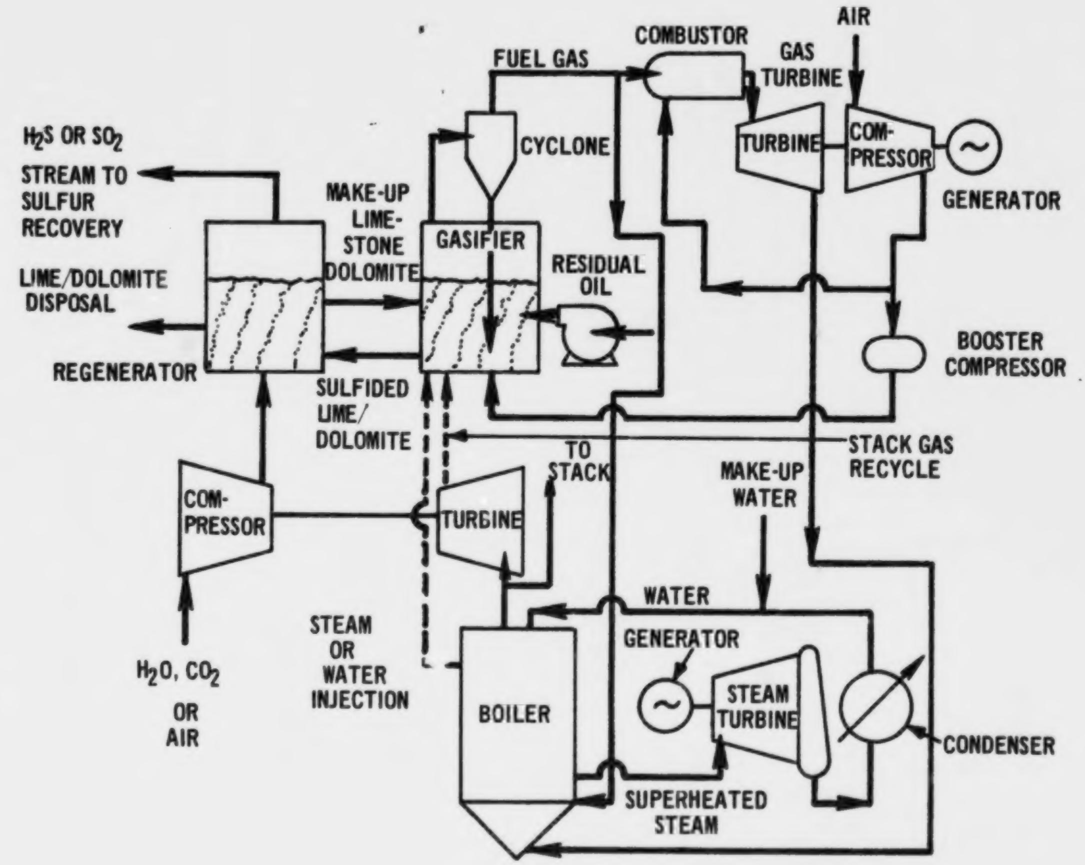

Figure 4. Regenerative pressurized oil gasification process.

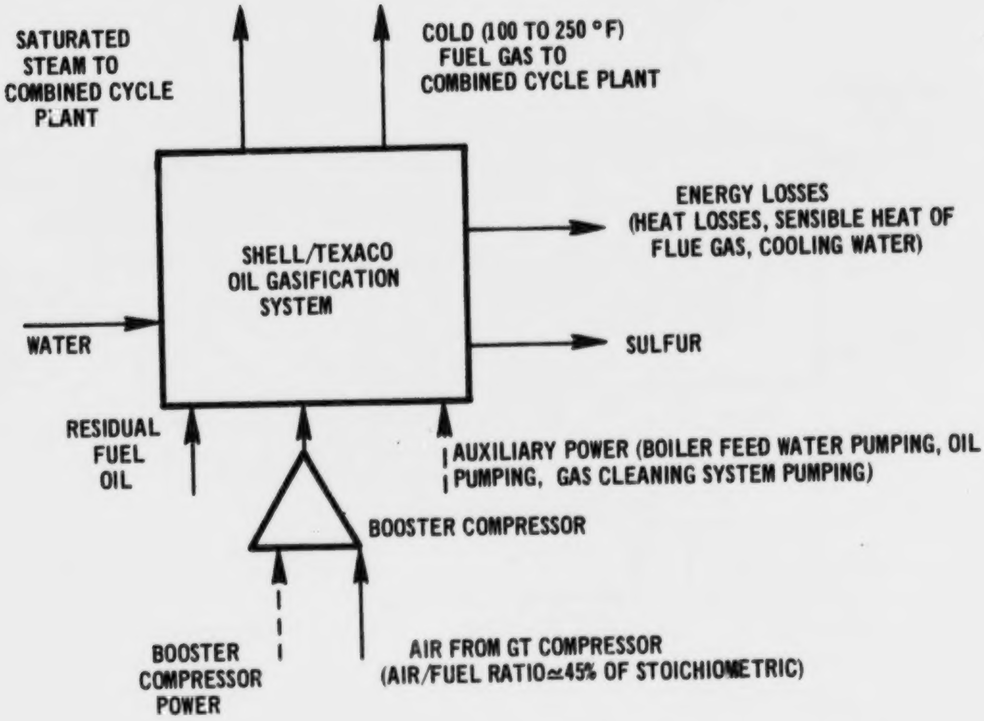
SATURATED STEAM TO COMBINED CYCLE PLANT
COLD (100 TO 250 °F) FUEL GAS TO COMBINED CYCLE PLANT
ENERGY LOSSES (HEAT LOSSES, SENSIBLE HEAT OF FLUE GAS, COOLING WATER)
SHELL/TEXACO OIL GASIFICATION SYSTEM
WATER
SULFUR
RESIDUAL FUEL OIL
AUXILIARY POWER (BOILER FEED WATER PUMPING, OIL PUMPING, GAS CLEANING SYSTEM PUMPING)
BOOSTER COMPRESSOR
BOOSTER COMPRESSOR POWER
AIR FROM GT COMPRESSOR (AIR/FUEL RATIO≃45% OF STOICHIOMETRIC)

Figure 5. Energy balance for the Shell/Texaco oil gasification process.

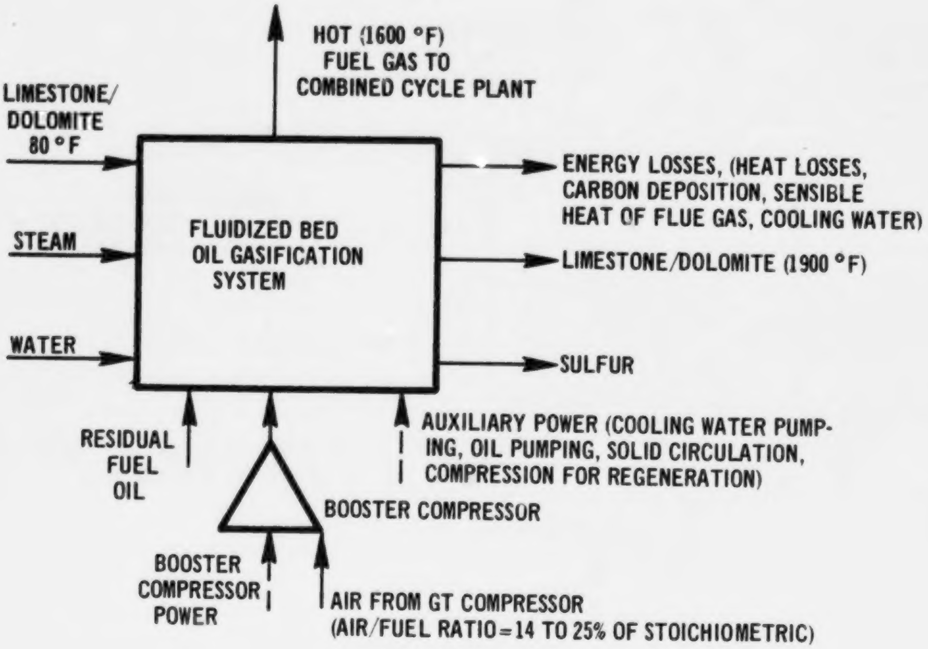
HOT (1600 °F) FUEL GAS TO COMBINED CYCLE PLANT
LIMESTONE/ DOLOMITE 80 °F
STEAM
WATER
FLUIDIZED BED OIL GASIFICATION SYSTEM
ENERGY LOSSES, (HEAT LOSSES, CARBON DEPOSITION, SENSIBLE HEAT OF FLUE GAS, COOLING WATER)
LIMESTONE/DOLOMITE (1900 °F)
SULFUR
RESIDUAL FUEL OIL
AUXILIARY POWER (COOLING WATER PUMPING, OIL PUMPING, SOLID CIRCULATION, COMPRESSION FOR REGENERATION)
BOOSTER COMPRESSOR
BOOSTER COMPRESSOR POWER
AIR FROM GT COMPRESSOR (AIR/FUEL RATIO=14 TO 25% OF STOICHIOMETRIC)

Figure 6. Energy balance for the fluidized-bed oil gasification process.

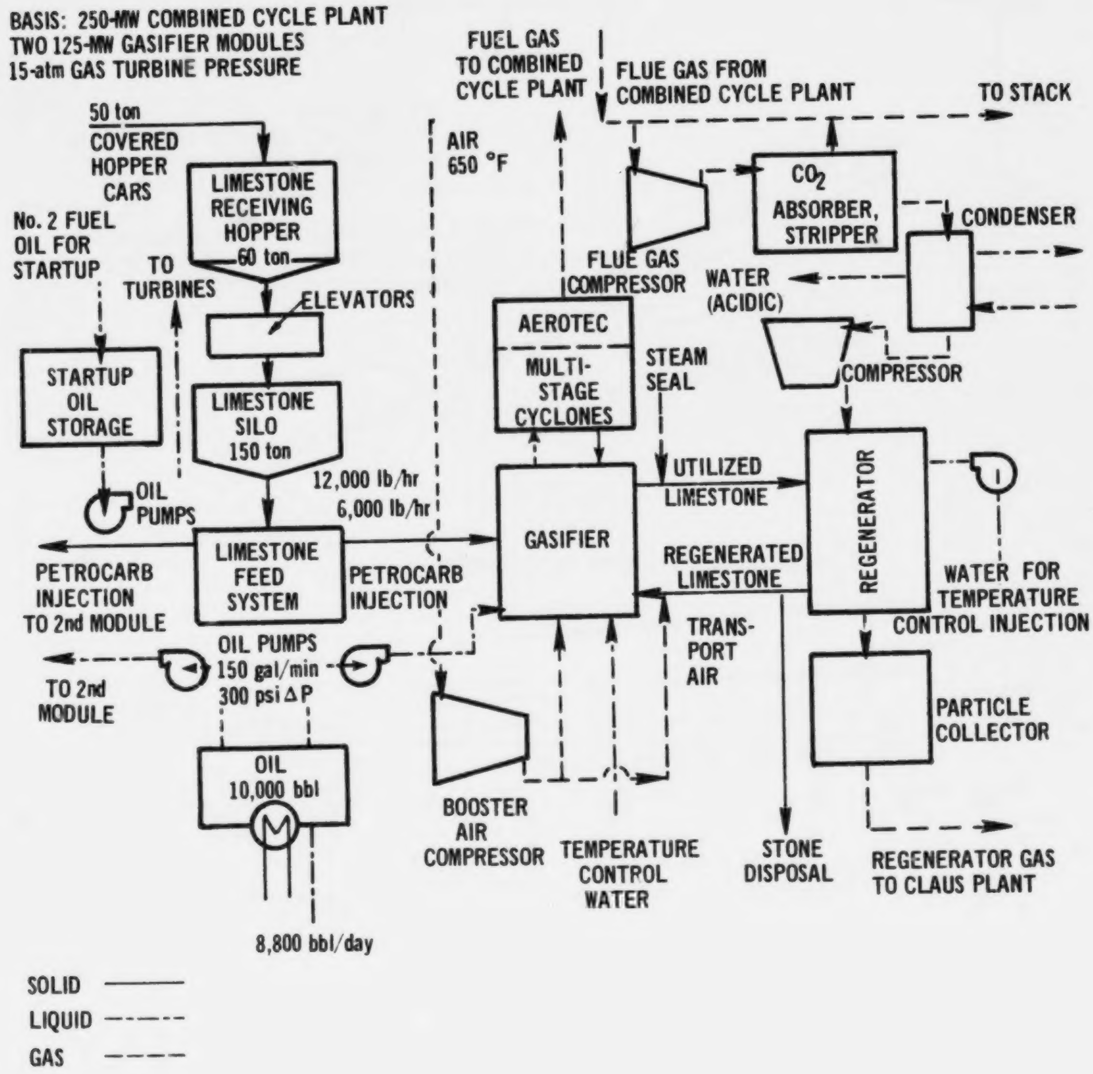

Figure 7. Pressurized oil gasification plant flow diagram.

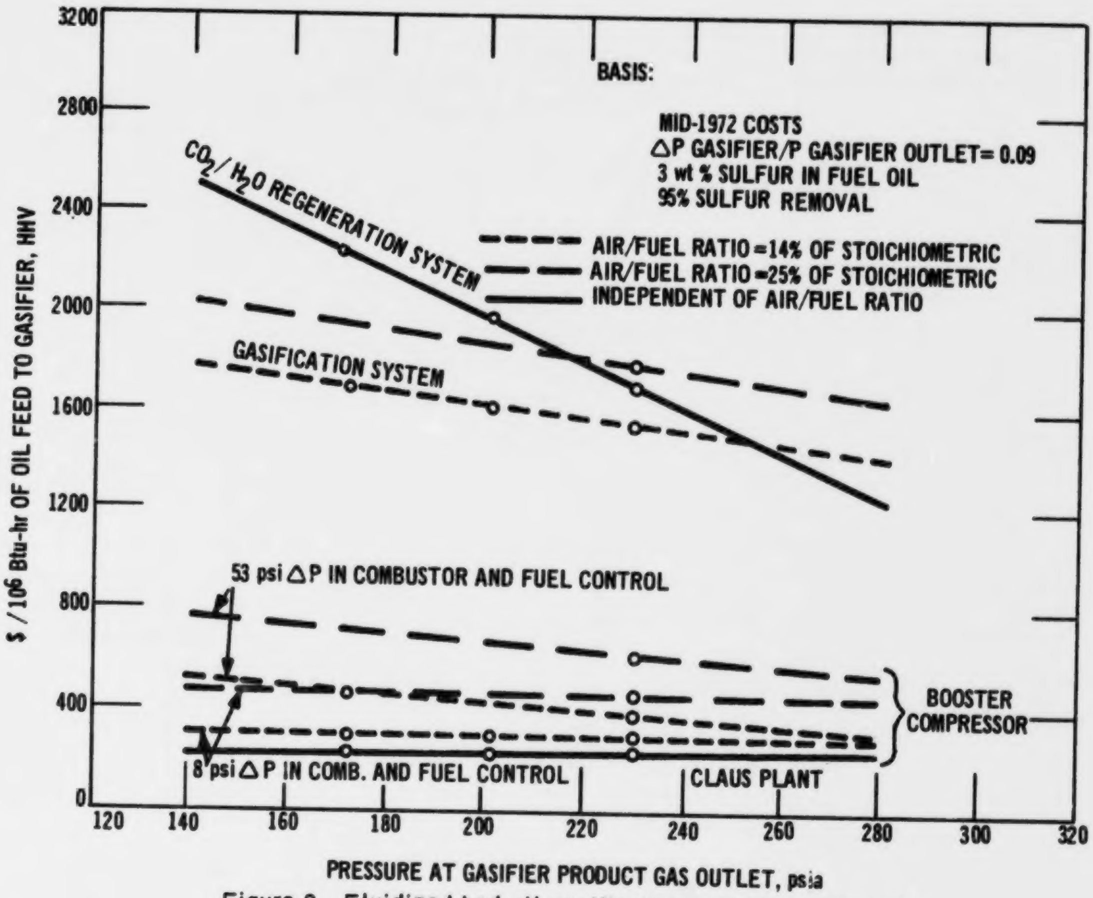

Figure 8. Fluidized-bed oil gasification capital investment.

6. FUEL GASIFICATION AND ADVANCED POWER CYCLES—A ROUTE TO CLEAN POWER

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ABSTRACT

The United States is currently faced with a growing gap between the demand for electrical energy and the supply of economic fuels for generating this energy with minimum environmental impact. The use of advanced power cycles utilizing technological spinoffs from the aerospace industry in conjunction with fuel gasification/desulfurization offers a solution which could prove to be not only technically feasible but economically attractive. A review of one such system, the Combined Gas And Steam (COGAS), is presented and the technical and economic advantages are enumerated. There are, however, several problem areas, particularly in the interface between the power system and the fuel system which must be resolved before the overall concept becomes a commercially viable one. These problem areas are presented with the intent of provoking thoughtful discussion and perhaps of opening new areas of research among the conference attendees.

INTRODUCTION

The majority of the people attending this conference were aware of the current energy crisis facing this country well before there was in fact a crisis. This is not the time to talk of the reasons for the current situation but rather to discuss methods of alleviating it by using the Nation's vast supply of coal, a fuel source now held in low esteem by a large segment of the air pollution regulatory agencies. It is apparent that unless extensive effort is directly applied towards this goal, or unless equally extensive institutional changes are brought about in this country, we will be faced with a utility system based upon foreign sources for one portion of our fuel and what can only be termed an adolescent nuclear industry for the remainder.

The use of fossil fuels in the utility industry will hopefully not parallel that currently being followed in the transportation industry where each new reduction in emissions is accompanied by a corresponding increase in fuel consumption. To assure this, any power system utilizing the advantages promised by fluidized-bed combustion/gasification must have the potential of achieving operating efficiencies significantly higher than currently attainable in conventional boilers.

However, the various economic forces, natural and imposed, which now and in the future affect the energy scene are such that a delay in the introduction of these advanced power systems could result in electrical power becoming more of a luxury than a necessity. Thus, these advanced power systems must be based upon technology which is now well in hand, but which will continue to grow, thus affording better performance and economics as each new generation of power system is achieved.

The system described briefly in the following paragraphs does indeed use the current technology available in the aircraft gas turbine industry applied to the industrial segment to realize efficiency and economic gains. Second and third generation power systems using evolutionary changes of technology currently being demonstrated will offer power systems which could make use of the pollution reductions offered by fluidized-bed combustors and still attain efficiencies well beyond those of conventional steam-electric plants.

THE POWER SYSTEM

Current steam power plants have efficiencies approaching 40 percent at 1000°F, a value which is limited not by thermodynamics but by economics. There have been steam systems designed and operated at temperatures of 1200°F, but the initial boiler and turbine costs were high and the maintenance associated with operation at this temperature eventually caused derating to 1000°F.[1] What, then, are the alternatives available to increase the efficiency of power systems? The answer comes of course from Carnot's law which defines the efficiency limit of any heat engine operating between two temperature limits. Referring to Figure 1, it can be seen that while the theoretical Carnot efficiency is well above that obtained by a real power cycle, several of the advanced power cycles demonstrate efficiencies which are nearly 70 percent of the theoretical limit. The three fossil fuel-fired sytems having the highest efficiency are based on the potassium topping cycle, the COGAS cycle, and an MHD topping cycle. Each of these cycles uses a high-temperature cycle to top a more or less conventional steam cycle. Of the three mentioned, however, only the COGAS system has been demonstrated in commercial size and, in fact, nearly 2500-MW of these systems are currently on order by the utilities.[2]

The advantage the COGAS system has over the potassium and MHD topping systems is that of development. The COGAS system is evolutionary in nature, adapting the advances demonstrated in military and commercial aircraft to the large industrial turbomachines. Thus, the technology of the JT9D engines, used on the 747, which allows operation at 2000°F and above will be adapted to the next generation of industrial machines.

In order to estimate the performance of the COGAS system, the relation of system elements must be established. While there are many ways of combining the gas turbine and steam system equipment, essentially two generic types emerge: (1) the waste-heat recovery type in which the turbine exhaust raises steam with or without additional firing, and (2) the pressurized-boiler type in which steam is raised and superheated using heat from a gas turbine combustor. These variations are shown in Figures 2 and 3, respectively. The effect of configuration of performance can be obtained by interpreting Figure 4, which shows station efficiency versus gas turbine participation for the waste-heat recovery type configuration.

The parameter gas turbine participation is really a measure of oxygen utilization. For a given turbine inlet temperature, a fixed amount of oxygen is required for combustion; the remainder, plus a large amount in the dilution air, is exhausted through the turbine. In the waste-heat recovery system, if there is no additional firing, the efficiency is the highest value, i.e., the right end of the lines of efficiency in Figure 4. If there is firing using up the additional oxygen and generating more steam, the gas turbine participation declines, and the efficiency becomes lower until the left end of the line, the point of which all the oxygen is consumed, is reached. The foregoing applies to combined-cycle systems with turbines operating at 2000°F and above. Below this turbine inlet temperature, the combined-cycle efficiency could, in fact, increase with high steam fractions since it is possible to have steam-cycle efficiencies significantly better than the efficiency of the lower temperature gas turbine.

In the supercharged cycle, fuel and air are burned essentially at stoichiometric conditions and the combustor exhaust is cooled to turbine inlet temperature by raising steam rather than be dilution air. This raising of steam by combustion has the same effect as supplementary firing, i.e. the system efficiency is reduced. Actually these systems are, obeying Carnot's law, which in essence says that the system which uses the entire heat input at the combined-cycle efficiency will be more efficient than the system which utilizes only part of the heat input at combined-cycle efficiency and part at the lower steam-cycle efficiency.

There are also considerations outside of the power cycle which influence the ratio of gas turbine power to steam power which will appear again when the integrated gasification/power system is discussed.

THE GASIFICATION SYSTEM

There is a wide variety of methods for converting solid or liquid fuels to gaseous form. Rather than discuss the operational characteristics of any one of these processes, it would prove more fruitful to discuss those characteristics which are important from the viewpoint of utilizing the fuel in an advanced-cycle power system.

Perhaps the most important aspect of fuel supply is cleanliness. While the turbine can handle a wide variety of fuels, each of these fuels must meet rather rigid contamination criteria. For gaseous fuels, the current specifications[3] will allow no more than 0.08 $lb/10^6 ft^3$ of total solids. No particulate size is specified, but filters are located in the fuel lines capable of removing particles of 30 μm and above. To minimize blade erosion, however, particulates should be small enough to follow the air stream through the blading without impingment. The exact particulate size has not been firmly defined but measurements of smoke particles seem to indicate few larger than 20 to 25 μm.

Current specifications limit total sulfur content in the fuel to 162 $lb/10^6 ft^3$ of which H_2S can be no more than 0.18 percent by volume. This amount, assuming it was all converted by combustion to SO_2, would be the equivalent of about 3 $lb/10^6$ Btu of methane-type fuel gas or about 2 $lb/10^6$ Btu of low-heating-value gas from coal. Both of these values are above EPA regulations (no allowable SO_2 for gaseous fuel, 1.2 $lb/10^6$ Btu for coal) so that any sulfur removal methods which will meet EPA regulations could be suitable from the turbine viewpoint.

A gas turbine can handle a wide latitude of fuel heating values, ranging from blast furnace gas ($\sim$ 100 Btu/ft^3) to propane ($\sim$ 2000 Btu/ft^3). Thus, the chemical heating value, per se, is not a problem. There is, however, a heating value dependent problem which must be considered. This is the problem of fuel delivery pressure. The first aspect of this problem is essentially one of hardware. The sizing of fuel manifolding, injection nozzles, etc., is a function of fuel heating value, i.e., a given Btu/min must be supplied to the engine and a fuel with a heating value of 150 Btu/ft^3 requires a higher volume throughput than one with 1000 Btu/ft^3. Thus, to reduce the fuel handling equipment to sizes compatible with high release gas turbine combustors, the low-heating-value fuels should be supplied at a pressure higher than the pressure ratio of the engine. The relationship between the fuel delivery pressure and the Btu/ft^3 requirement differs among engine types but is of the form: $P = (f)\ LHV/(\text{Specific gravity} \times \text{delivery temperature})^{1/2}$. Unfortunately the functional form is inverse; thus, as the heating value decreases or temperature increases, the fuel delivery pressure increases. This has a significant effect on overall system performance and will be discussed later in the paragraphs dealing with the integrated station performance.

The most important characteristic of a gasifier designated for use with a power system is its efficiency in converting coal Btu's to fuel gas Btu's. There are two gasifier efficiencies that need to be considered. The first is the cold gas efficiency (chemical heating value of fuel gas/chemical heating value of coal) and the second is the hot gas efficiency (chemical & sensible heat in fuel gas/chemical heating value of coal). The manner in which these two efficiencies affect the integrated system performance is complex, but if one thinks again in terms of Carnot's law, all of the chemical heating value (represented by the cold gas efficiency) is used at combined-cycle efficiency, while the sensible heat may or may not be so utilized. If the hot gas can be used in the engine, there would be no degradation in the performance. However, if the gas must be cooled by raising steam, then the sensible heat could be used only at steam cycle efficiency. There is of course, a "however", attached to the foregoing. If the sensible heat were to be used in regenerating the gasification system, it would be used at essentially combined cycle efficiency. This use of the fuel gas sensible heat requires relatively expensive heat exchange equipment.

Another gasifier attribute necessary for use with power plants is operational flexibility. While power systems designed for base-load operation do not require fast startup capability, they do operate over a range of power settings ranging from perhaps 70 percent to full power. Thus, even base-load applications require some turndown capability. One of the more attractive features of the COGAS system is its capability of relatively fast startup, 5 minutes to full power for the gas turbine power and less than 1 hour for the total plant. This capability lends itself to applications in mid-range load factor (3000 to 6000 hr/yr) in which daily startup would be common place, with shutdown over weekends or holidays. A gasifier for use in this system would have to have a fast-start capability. Since this type of power system typically operates over a range of power settings from 40 percent to full power, flexibility in gasifier operation would be necessary.

INTEGRATED FUEL PROCESSING/ POWER SYSTEM

One of the basic tenets of mathematics is that the whole is equal to the sum of its parts. The power system has, in a sense, circumvented this by putting together two parts of comparable efficiency into a combined system of significantly better performance. It would be indeed fortunate if this symbiotic relationship could extend to the joining together of the fuel processing and power cycle portions into an integrated system. However, the serendipity does not carry over; in fact, the requirements at the interface of the two parts can cause a noticable reduction in combined performance. There is also an environmental consideration involving the production of NO_x which could influence the selection of overall systems configurations.

One of the simplest of all integrated systems is shown in Figure 5. Air for the gasifier is bled from the compressor, raised to the required gasifier pressure in a booster compressor, mixed with the fuel in the gasifier with the resultant hot fuel gas being supplied directly to the turbine. Some heat exchange between the air streams would be possible. This system utilizes the fuel sensible heat in the engine.

A much more complex configuration (Figure 6) results if the fuel sensible heat must be recovered for use elsewhere in the cycle. In Figure 6, the gasifier is run at essentially atmospheric pressure with the fuel gas exiting to a boiler/superheater. From there the fuel gas passes through a heat exchanger where bleed air from the compressor enroute to the gasifier is heated, then through a feed water heater and finally into the booster compressor were it is raised to the required pressure for injection into the gas turbine burner. The booster compressor could be driven, in part, by an expansion turbine in which the heated bleed air is let down to gasifier pressure. The feedwater and superheated steam would be utilized in the waste heat boiler.

What are the performance differences of these two systems? The simple systems utilize all the fuel heating value at combined-cycle efficiency, while the second utilizes only the chemical heating value at combined efficiencies with the sensible heat being used at steam cycle efficiencies. The efficiency differences can be found by inspecting Figure 7, based upon data from Reference 4, in which the percentage change in efficiency with fuel temperature is shown for three different turbine inlet temperatures. From all indications, the system utilizing the sensible heat in the gas turbine is the most efficient. This reinforces the results shown in Figure 2, which indicated that large gas turbine participation was more efficient.

There are, however, several considerations that must be made before a final choice can be made. Besides the very difficult hardware problems associated with fuel control systems handling gases at 35 to 50 atm and temperatures above 2000°F, there are the environmental constraints to be considered. The systems pictured in Figures 5 and 6 are based upon both sulfur and particulate removal within the gasifier; i.e., a method of desulfurization and particulate removal that operates at 1500°F and above. Currently, the majority of sulfur removal systems operate at 600°F or below, some even requiring below zero gas temperatures. With the exception of fluidized-bed gasifiers and perhaps one or two other types, the majority of gasifiers require external desulfurization and thus require some method of heat recovery from the fuel gas stream.

A second environmental constraint is that of NO_x formation. It is well established that the formation of NO_x is strongly dependent upon the combustion temperature. Combustion temperature, in turn, is a function of both fuel heating value and of fuel and air preheat. This dependency is shown in Figure 8 where it can be seen that the combustion temperature is a stronger function of sensible heat than of HHV. Using Figure 9, which shows NO_x concentrations as a function of temperature, the rise in combustion temperature due to fuel gas sensible heat will increase; in the extreme, the emission of NO_x by a factor of about 25 (i.e., from less than 5 to nearly 100 ppm with a fuel gas having an HHV of 120 Btu/ft^3). As the fuel HHV increases, the base NO_x emission factor increases and the multiplying factor due to sensible heat decreases — slowly at first and then rapidly as NO equilibrium is approached. The allowable concentration, using EPA regulations for NO_x from coal-fired power plants would be in the order of 120 to 160 ppm, depending on engine efficiency. (If the system were to be considered as a gas-fired station, the allowable concentrations could be 35 to 45 ppm. A brief discussion of emission regulations and their form is given in Appendix A.)

The situation, therefore, is that as the HHV of the product gas increases, the sensible heat must be decreased (or vice-versa) in order to meet the NO_x standards. Since the NO_x production increases more rapidly with sensible heat, it would seem to be more advantageous to have a gasifier with a high cold gas efficiency (more HHV in the gas) as was mentioned before. This is doubly beneficial since the HHV of the fuel also has a noticeable effect on the integrated system efficiency (Figure 10). (This effect is somewhat exaggerated in Figure 10 since the steam system efficiency used in preparing this figure was relatively low, i.e., ~30 percent.) This is because of two effects, a reduction in steam generation due to the decrease in mass flow of fuel, and a concurrent decrease in booster compressor work.

There are several ways to increase the HHV of the product gas. Two of the more promising are: (1) regeneration of the gasifier air, and (2) oxygen enrichment of gasifier air. In the first, the air to fuel ratio needed to attain a given gasifier temperature is reduced as the air is preheated. This means less nitrogen dilution and more Btu/ft^3. Oxygen enrichment accomplishes the same thing; i.e., a reduction in nitrogen dilution thereby increasing HHV. In fact, the use of oxygen alone to blow the gasifier would result in the production of synthesis gas having an HHV of about 315 Btu/ft^3. Unfortunately, the combustion of synthesis gas could result in greater NO_x production than the combustion of methane.

There are, or course, modifications which can be made to the combustion process which could effect a reduction in NO_x emission. One of these, off-stoichiometric combustion resembles, in theory, the method being used with some success in gas-fired steam boilers. At this time, there is no assurance that the combustion efficiency during this staged-combustion will be comparable to that currently obtained in gas turbines, e.g., greater than 99 percent. This is especially true with the low-Btu fuels whose combustion characteristics in gas turbine burners have not been extensively studied.

A SUMMING UP

The foregoing discussions have briefly described the advanced power system which offers the potential of attaining efficiencies nearly 50 percent greater than those currently obtainable yet utilizes equipment which would be evolutionary developments of the combined-cycle systems that are currently being placed on-line by utilities. When used in conjunction with coal/residual oil gasification, such systems could generate electrical power with a minimum of pollution while using relatively abundant high-sulfur fuels. Although economics have not been treated thus far, prior studies have indicated that the combined fuel gasification/advanced power system could generate this pollution-free power at costs less than conventional steam power plants having equivalent emission characteristics and at costs which could easily be less than those associated with nuclear power (Figure 11 from Reference 5).

That this answer to a maiden's prayer is not without problems is apparent even to the casual reader of this paper. Some of these problems have been touched on — configuration of the power cycle, pressurization level of the gasifier, trade-offs between sensible and chemical heating value, etc. A myriad of practical problems exist in interfacing the two complex systems.

However, many of these problem areas can be resolved only by construction and operation of a prototype plant of large enough scale to demonstrate the concept that clean power can be generated from high-sulfur fuels with acceptable economics. Until a successful demonstration(s) takes place, utilities will view the concept as just that—a concept—and will continue to displace fossil-fired systems with a greater dependence on nuclear power. The recent announcements by the Office of Coal Research describing its plans for one or more demonstration plants gives hope that a reasonable program will soon be underway — one which will lead to the introduction of commercial systems by the latter part of this decade.

REFERENCES

1. Giramonti, A. J. Discussions of Steam and COGAS Systems with the Babcock and Wilcox Co., Barberton, Ohio. UARL Report UAR-H246, 1969.
2. Packaging Sells the Combined Cycle. Electrical World. September 1, 1972.
3. P&WA Specification 526 - Gaseous Fuel. Industrial for Turbine Engine. June 1968.
4. Giramonti, A. J. Advanced Power Cycles for Connecticut Utility Systems, UARL Report L-971090-2, January 1972.
5. Robson, F. L. Clean Power from Gas Turbine-Based Utility Systems. Combustion. July 1972.

APPENDIX A

Emission Standards

Current emission standards are based upon the amount and type of fuel burned; x pounds/10^6 Btu for coal, y lb/10^6 Btu for oil, and z lb/10^6 Btu for gas. If the full advantage is to be taken of the advanced power systems, a new basis for standards must be used.

First, as was alluded to in the main text, fuel type should be defined in a different manner. The power system burns a gaseous fuel even though coal, residual oil, coke, or garbage is used in the gasifier. In fact, in an actual system it would be hoped that the input to the gasifier could be switched more or less as the fuel market dictates.

Secondly, the current standards are based upon input rather than the output, which is the real purpose of the power system. It is now possible for a power station to meet the regulations but emit, in absolute numbers, significantly more pollutants than would an advanced power system of equal power. For example, a conventional steam station of 1,000-MW output running at off-design conditions to meet NO_x regulations could have a heat rate of perhaps 12,000 Btu/kWhr and, meeting EPA standards, would put out

2,400 lb/hr or 2.4 lb/MWhr of NO_x. An advanced system having a heat rate of 8,000 Btu/kWhr and meeting the same standards as currently written would emit, for the same power demand, 1,600 lb/hr or 1.6 lb/MWhr of NO_x.

As turbine inlet temperatures increase, there is a second-order increase in NO_x emissions. While minor, this increase could result in an emission/10^6 Btu above the prescribed level. However, the power system has become more efficient, and there could well be a decrease in emissions/output compared to the lower temperature system. Referring to the above example, suppose a 500°F increase in turbine inlet temperature caused a 10 percent increase in NO_x emissions but also a 20 percent decrease in heat rate. The system would no longer meet the current EPA regulations but would, in fact, emit only 1.4 lb/MWhr.

For these reasons it appears that standards based upon emissions/MWhr would be a more reasonable choice.

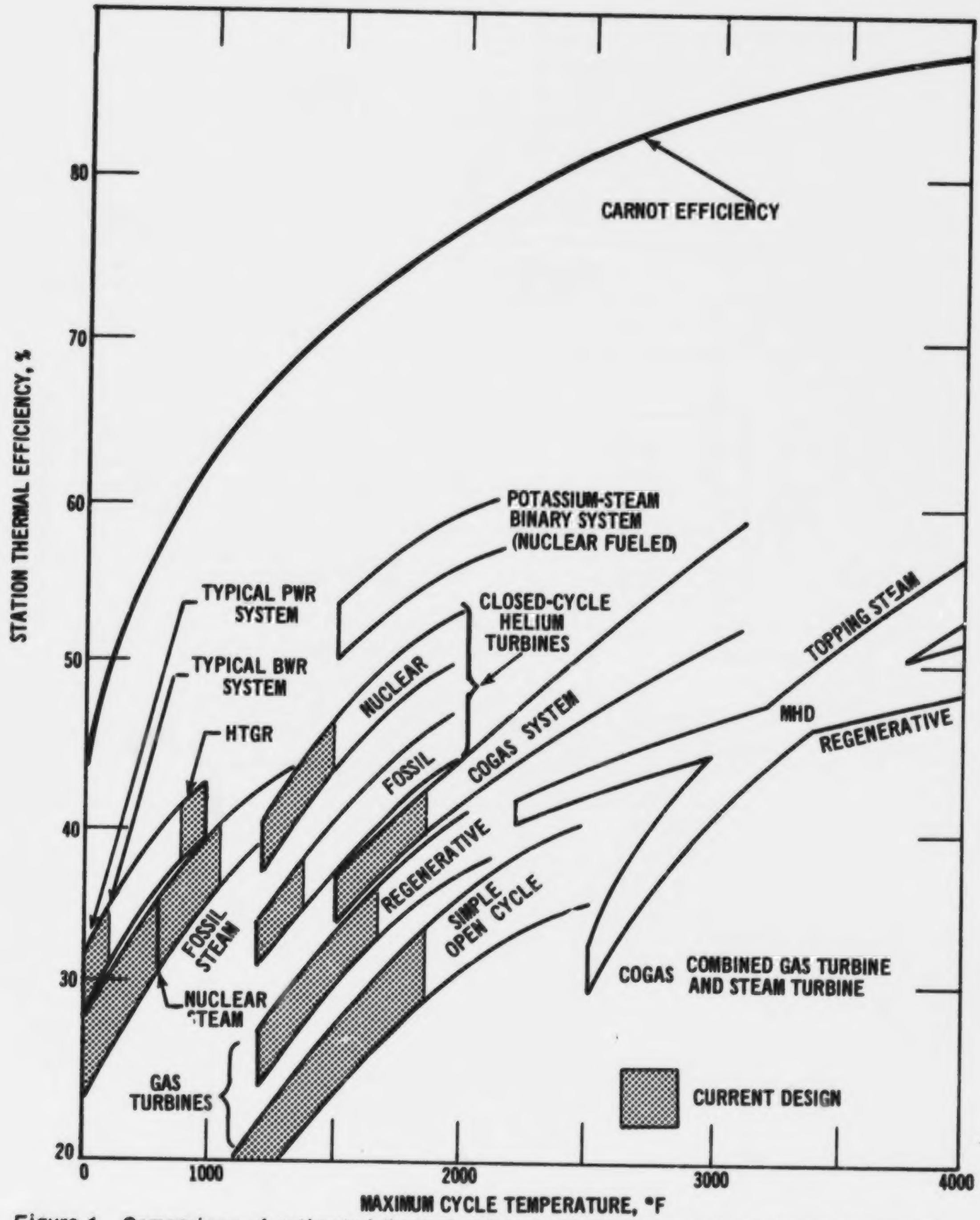

Figure 1. Comparison of estimated thermal efficiencies for advanced-cycle power stations.

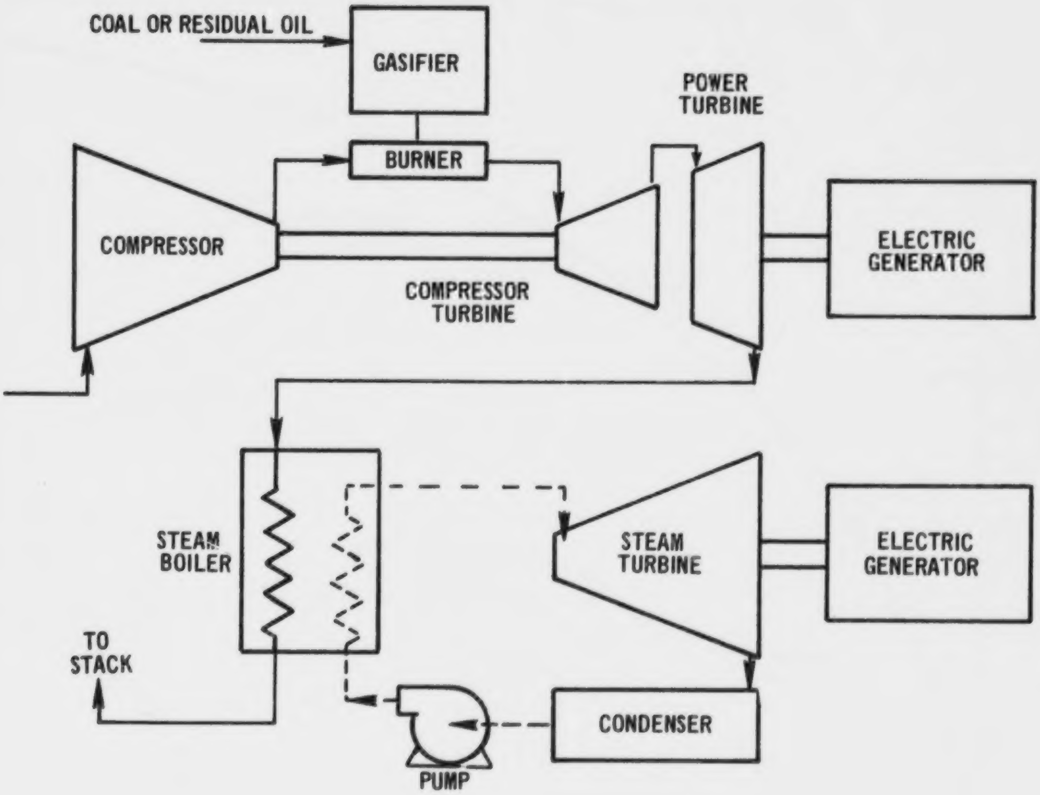

Figure 2. Combined gas-steam turbine system.

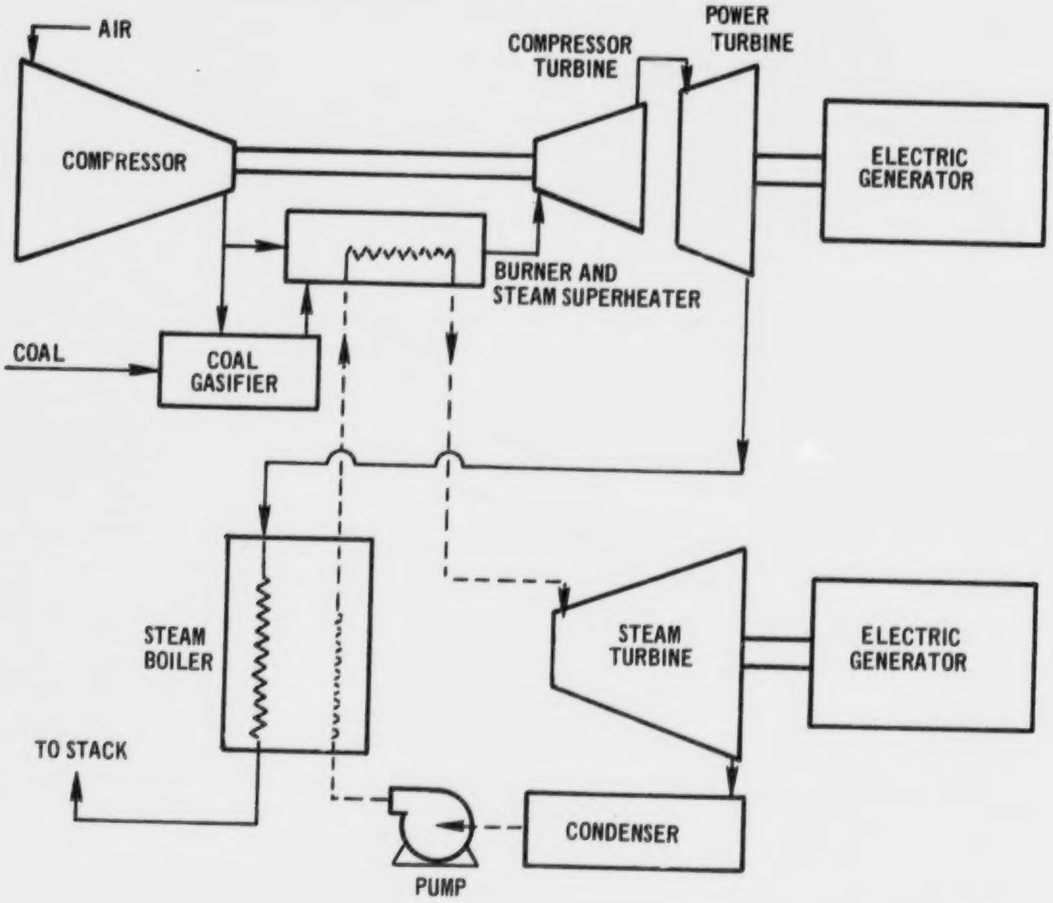

Figure 3. Supercharged combined gas and steam turbine system.

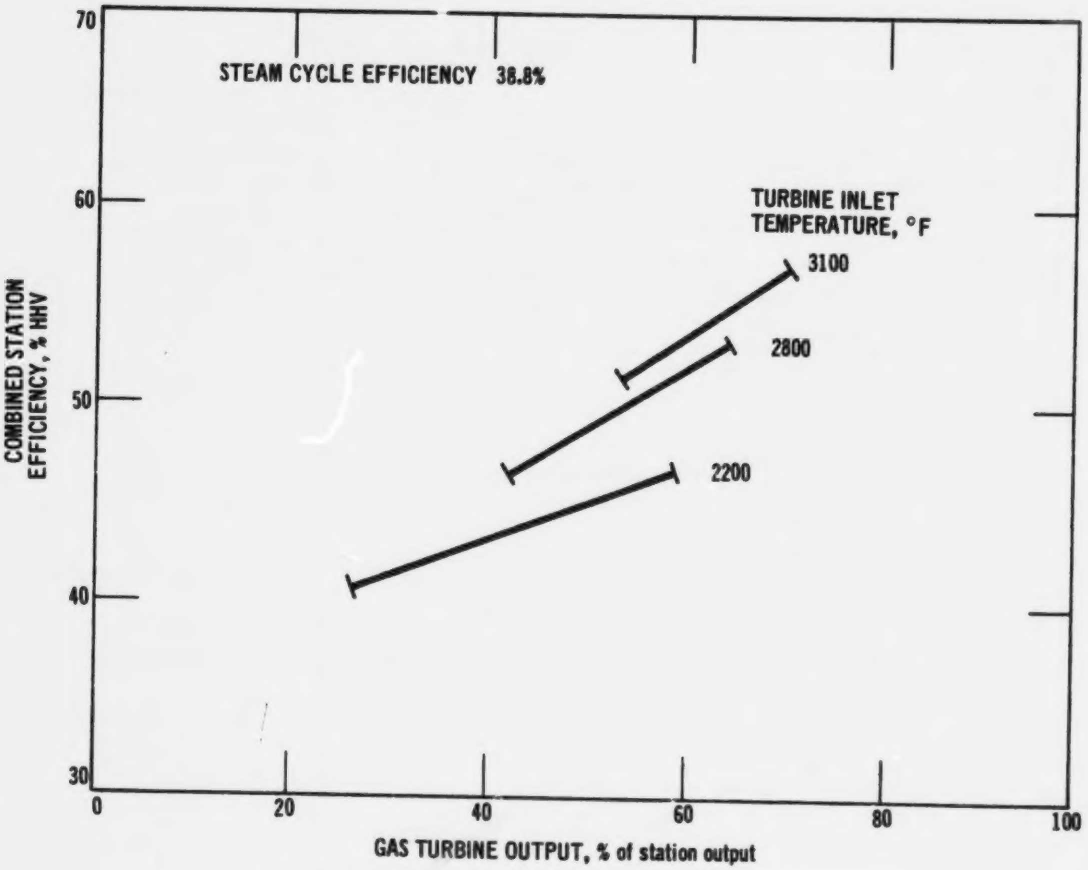

Figure 4. Performance of exhaust-fired combined system.

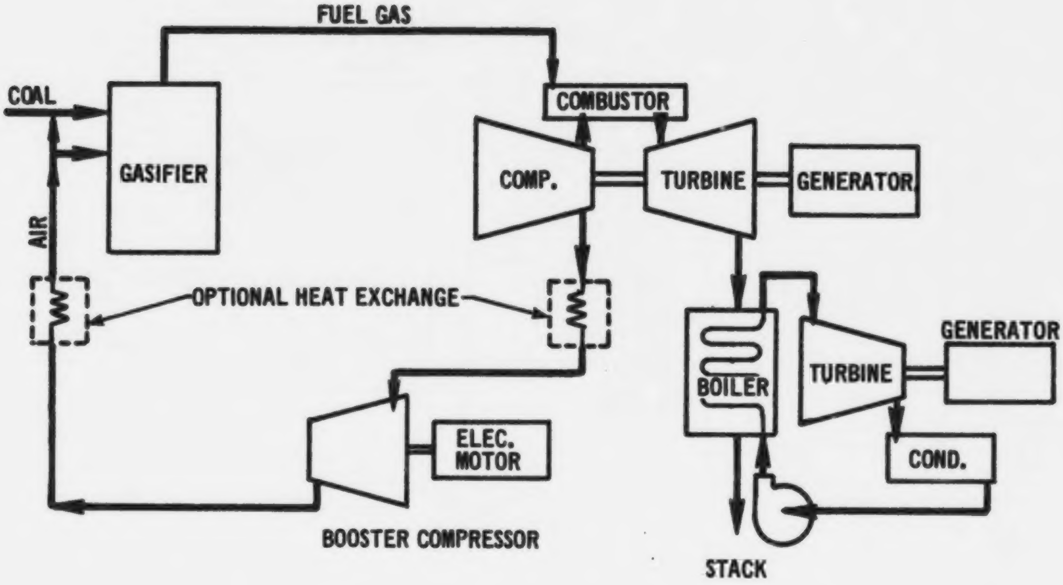

Figure 5. Schematic of high-pressure system.

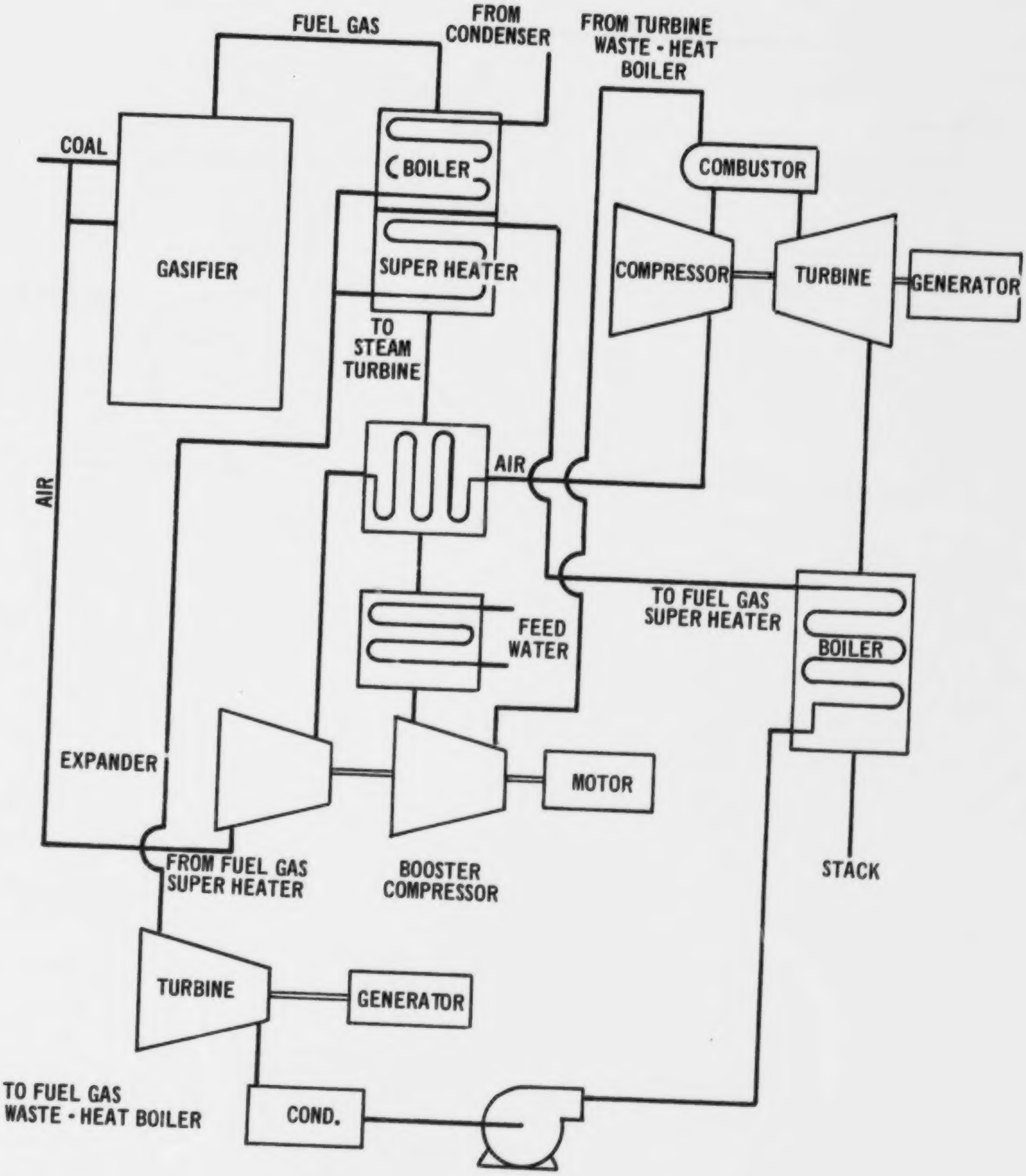

Figure 6. Schematic of low-pressure system.

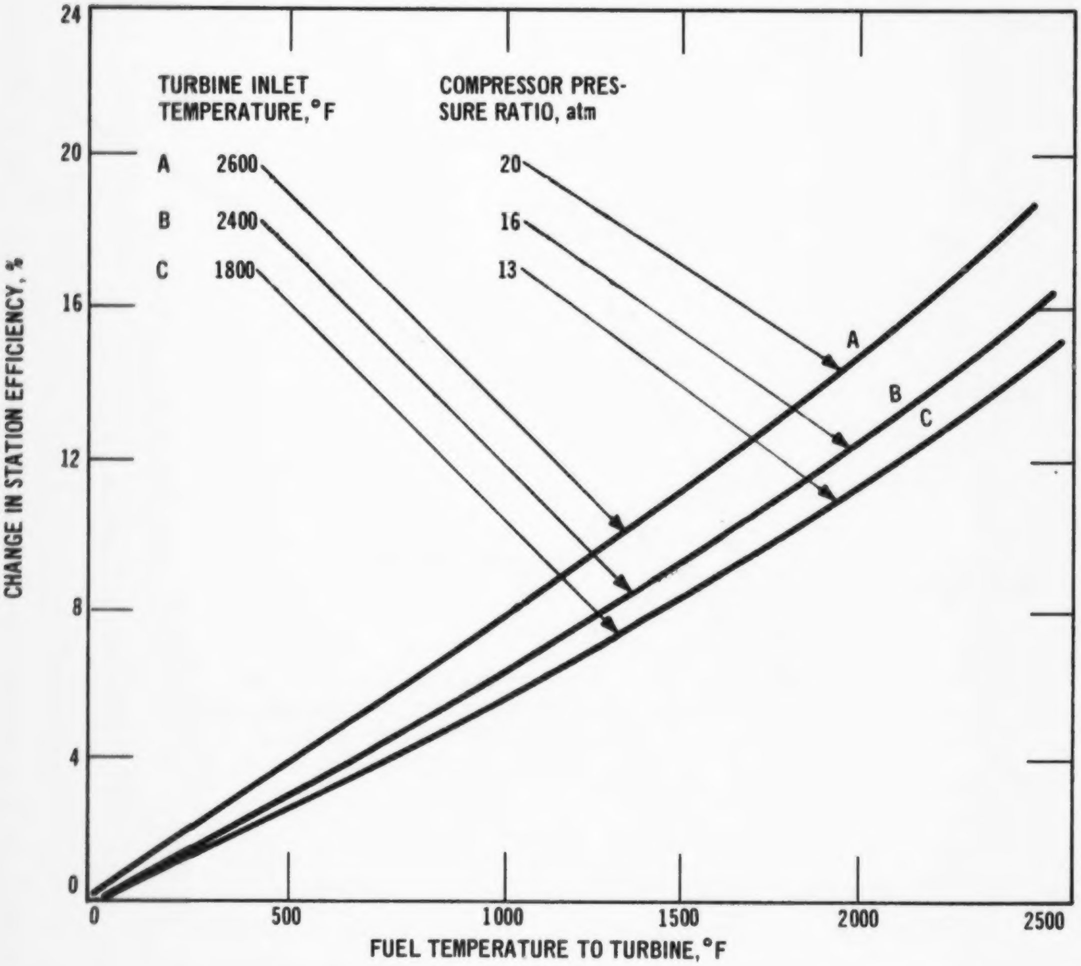

Figure 7. Effect of fuel temperature on integrated station efficiency.

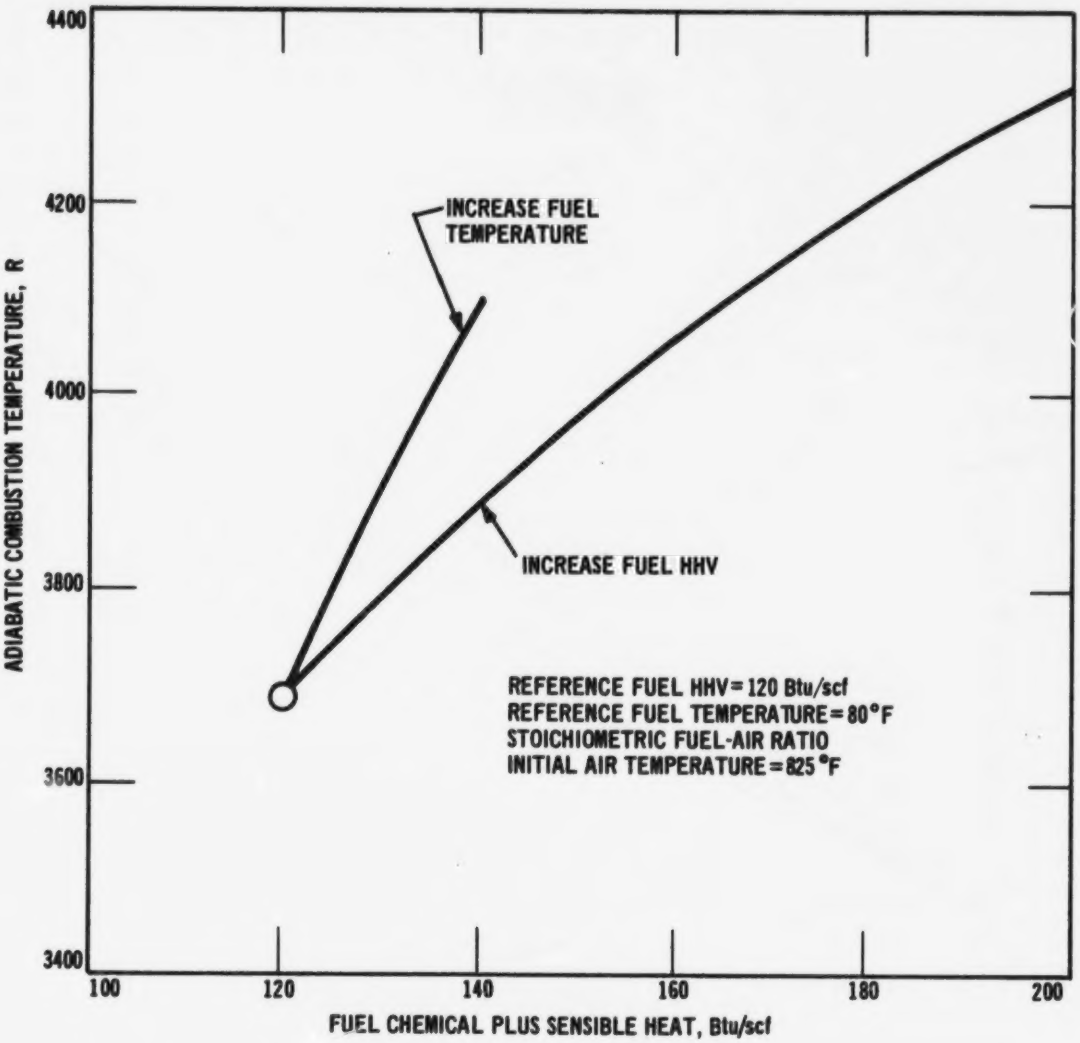

Figure 8. Effect of fuel gas chemical and sensible heat on combustion temperature.

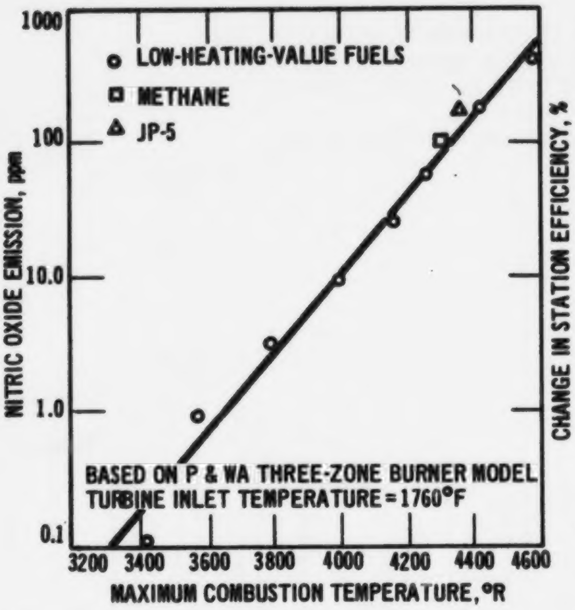

Figure 9. Effect of combustion temperature on nitric oxide emissions.

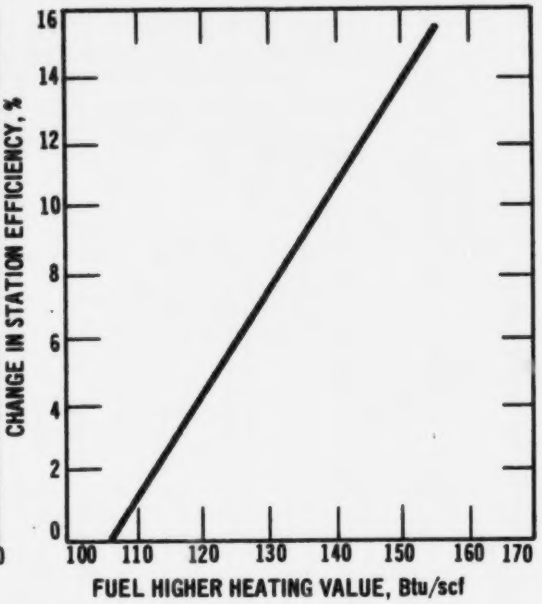

Figure 10. Effect of fuel HHV on integrated station efficiency.

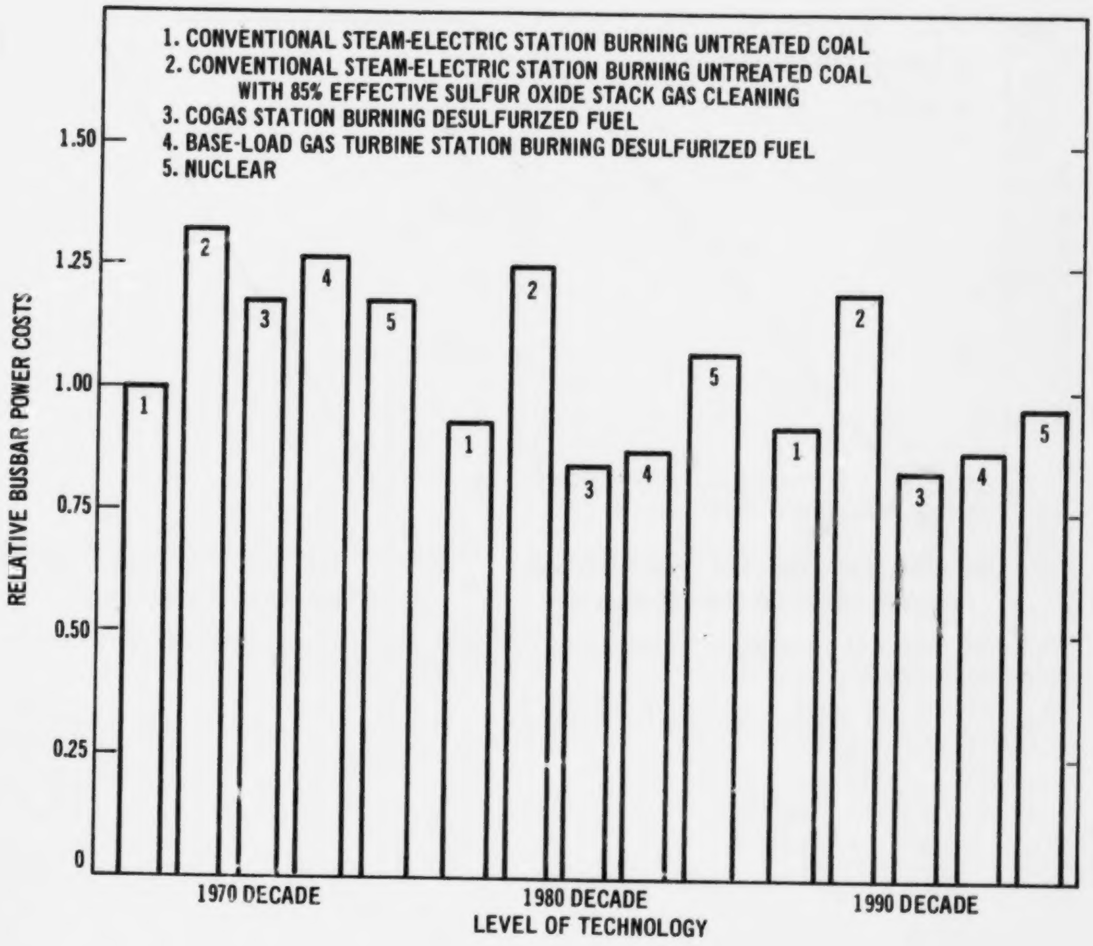

Figure 11. Busbar power costs for coal-based power plants.

7. A DESIGN BASIS FOR UTILITY GAS FROM COAL

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INTRODUCTION

The preferred solution for control of atmospheric pollutants from the stacks of electric utility boilers is to substitute clean fuel for polluting fuel. In many cases a clean fossil fuel such as natural gas is not a practical substitute for coal in the generation of electricity because of scarcity and cost. Furthermore, the available supply of clean fuel may combat pollution more effectively when used to fulfill residential and small commercial needs.

The combustion products of coal contribute one-eighth of the total atmospheric pollutants emitted in the United States, including one-half of the sulfur oxides and one-quarter of both the nitrogen oxides and of the particulates. Sulfur emissions from coal combustion may be reduced by: (1) using low-sulfur coal, (2) cleaning high-sulfur coal by physical methods, (3) removing sulfur oxides from coal combustion gases, (4) removing sulfur during the combustion step, (5) producing de-ashed low-sulfur fuel by solvent processing of coal, and (6) gasifying coal and removing sulfur from the gas before combustion.

The last method, coal gasification with gas cleaning before combustion, promises the greatest reduction in sulfur emission. Most of the sulfur gasified appears as hydrogen sulfide. Several different commercial gas cleaning processes are available today which can reduce the hydrogen sulfide content of gas streams to less than 10 ppm; some processes can remove hydrogen sulfide to 1 ppm or less.

This paper discusses the selection of design criteria for the gasification of coal and cleaning of the generated gas before combustion in an electric utility boiler. The preliminary plant design description is for a large pilot plant installation that will demonstrate the feasibility of this concept.

The gas produced from coal for the boiler is called utility gas, although producer gas and low-Btu gas are equivalent names. It is made by gasifying coal with air and steam at elevated pressure. Dust and sulfur compounds are removed from the utility gas before it is burned in the power generating system. Heating value of the utility gas will be between 140 and 250 Btu/ft^3, depending on the gasifier and the plant design.

The low heating value of utility gas limits the distance it can be transported economically. When used to fuel electric power stations, it probably will be generated onsite. Retrofitting coal gasifiers to existing boilers is one of the most important applications now for coal gasification plants. In the future installations, combined gas turbine-steam turbine systems will be served by gasification plants not only for reduced atmospheric pollution, but also for greater efficiency in the generation of electricity. From these plants we expect savings in investment and decreased electricity costs as well as less heat rejection to the environment and better conservation of our coal resources.

We believe that this country has an urgent interest in the demonstration of the practicability of coal conversion to clean utility gas for the electric power industry as soon as possible. Successful achievement of this goal in the shortest possible time through government and industry support will provide substantial benefits to the country and to the electric utility industry.

The program proposed by the Institute of Gas Technology (IGT) for proving this concept includes construction in the near future of a large pilot plant which will be located near an existing boiler. The pilot plant will be capable of feeding from 10 to 50 ton/hr of coal and will fuel a power plant with a generation capacity of 20 MW or more. We believe that with favorable results from this pilot plant commercial plant designs can be undertaken by the end of 1975.

GENERAL DESIGN PRINCIPLES

The entire concept hinges on the coal gasifier performance. The gasifier operation must be reliable; it must gasify a high percentage of feed carbon; and it must be load-following, that is, capable of operating in response to the power system requirements. The focal point of our effort is to demonstrate gasifier operation.

We anticipate that coal conversion to utility gas will be practiced to a great extent in the coal belt from Illinois to New York. Because the coals from this region have caking properties, the plant must be capable of accepting these coals as feed.

Initially, utility gas will be produced for existing boiler systems. Operation of the large pilot plant facility will show the practicality of retrofitting coal gasifiers to existing boilers. This is one of the most important applications now for coal gasification plants. In the future, for savings in investment and for decreased electricity costs, combined-cycle systems will be served by gasification plants. Flexibility will be provided in the pilot plant design to test advanced power generating system components.

The pilot plant gasifier will be large enough so that scale-up to commercial-size gasifiers in the future can be done with confidence. For example, a single gasifier fed at a rate of 10 ton/hr and operating at 300 psia will fuel a power station generating about 20 MW of power. Operation at 1000 psia will increase its capacity by more than 3 times. The diameter of this gasifier permits shop fabrication and rail shipment for reduced construction time and cost.

Our guiding principles in design of the first large pilot plant for conversion of coal to utility gas are:

1. Prove the gasifier design and operation.
2. Accept caking coal as feed.
3. Demonstrate application of coal gasification to existing boilers.
4. Build gasifier large enough so that it can be directly scaled to commercial size.

UTILITY GAS FROM COAL PLANT

Figure 1 is a block flow diagram for our proposed utility gas from coal plant. This design is suitable when the gasification plant is installed to fuel a small to medium size boiler. I will briefly describe the flow scheme in Figure 1 and then discuss in more detail the important parts of the plant.

The coal feed is crushed to the desired size. Lock hoppers are used to transfer coal from atmospheric pressure to the elevated pressure of the gasifier. Heat is recovered from the hot raw gases, and a small part of the cooled gas is used to pressurize the lock hoppers. The main gas stream expands from high to low pressure through a gas expander, thereby generating power needed to drive the large gasifier air compressor. Gas vented from the lock hopper system rejoins the low-pressure main gas stream. The combined gas streams are cleaned of sulfur at low pressure by the Stretford process or one that is similar. The Stretford process produces elemental sulfur directly from hydrogen sulfide. After sulfur removal, the gas flows to the boiler. Associated with this process train are a large air compressor, ash handling and disposal equipment, waste-water treatment, and possibly oil stabilization and storage equipment. When an efficient, high-temperature sulfur removal system has been developed, we believe that it will replace the equipment shown within the dashed lines.

DESIGN PRESSURE

In plants manufacturing pipeline gas from coal, maximum methane formation within the gasifier is desired for improved thermal efficiency. To obtain this, gasifier pressures of 1000 psia or more are preferred. The thermal efficiency of utility gas plants does not depend on methane formation within the gasifier, and, therefore, we have more flexibility in selection of plant pressure. If necessary — and this is what we have done — the plant pressure is established on other considerations than the chemistry of gasification.

Lock hoppers were selected to transfer coal into the plant. A dry solid feed permits a less complicated gasifier design, and the plant following the gasifier is also less complicated. For high plant reliability when using dry feed systems, the performance of lock hopper valves will probably set the upper pressure limit. Today, the best commercial lock hopper operation is that demonstrated with Lurgi gasifiers. In these, the pressure difference between lock hopper and gasifier is about 300 to 350 psi. Even though we want to gasify at pressures up to 1000 psi, we will design for a pressure of about 300 psi because lock hopper valves are available which work at this pressure. We intend to search for improved methods to feed dry coal into higher pressure systems.

COAL FEEDING SYSTEM

Figure 2 presents a simplified illustration of the solids handling system, which includes the coal feed system, pretreatment, gasification, ash removal, and dust removal from the gas.

As described above, the single-stage lock hopper was selected to transfer coal from atmospheric pressure to the elevated pressure of the gasifier. This feed system was chosen so that the gasification plant will be simple, reliable, and less costly.

The disadvantages of the lock hopper are evident. The most important is the difficulty in obtaining reliable operation of the lock hopper valves. During the hopper cycle, these valves alternately seal against the gasifier pressure, and then open to pass a fine, dry, abrasive solid. Coal dust tends to pack in the valve, cut the packing, and jam in the guides and seat. The valve must seal fairly well. Leakage overheats the valve and lock hopper; introduces dirty, raw gases into the hopper and its vent gas system; and wets the cool coal with moisture, oil, and tar causing the coal to bridge and no longer flow easily.

A second disadvantage is the need to vent gas from the hopper during its operating cycle. Loss of this vent gas decreases process efficiency. Recompressing the gas into the system is expensive. In this utility gas plant design, the vented gas is collected and mixed with the low-pressure main gas stream before sulfur removal. In case the valves leak, the vent gas system includes a cooler and vapor-liquid separator to reduce contamination of product gas.

The IGT HYGAS process is designed to manufacture pipeline-quality gas from coal. In this process coal is fed to the gasifier in the form of a slurry which is pumped to system pressure. This is a more reliable feeding system than high-pressure lock hoppers, and the slurry pumps are capable of good operation to 1000 psi and higher discharge. You might ask: Why not use a slurry feed system for utility gas generation?

Slurry feeding introduces additional complexity in the plant. Equipment for making the slurry must be provided. When the slurry enters the gasifier, the liquid must be vaporized. Therefore a drying bed is added to the gasifier, and the hot, raw gasifier gases are used to supply the heat needed for drying and for stripping. The lowered gas temperature makes efficient recovery of heat from the gas difficult. Stripped slurry oil must be recovered efficiently for recycling. In its recovery, more than one stage of quenching may be needed; the gas stream is cooled to 100°F, thus adding to the cooling water demand; and activated carbon towers or sponge oil scrubbing completes final oil removal from the gas. Then, the recovered oil has to be dewatered and stripped of dissolved gas before returning to the slurry tanks.

We concluded that utility gas plants for the electric power industry will be less complex, less costly, easier to operate, and more efficient when using lock hoppers to feed coal into the gasifier.

COAL PRETREATMENT

Most bituminous coals have the property of caking or agglomerating when heated. Agglomeration of coal within the gasifier cannot be tolerated because of the possibility of pugging. So that the utility gas process can accept the widest variety of coal as feed, facilities for modifying or destroying this property must be a part of the process.

Pretreatment takes place in the presence of air at 750 to 800°F. The particle surfaces of coal are mildly oxidized, destroying the caking properties. Heat is evolved and must be removed to control the temperature. Pretreated char yield is about 90 percent of the coal feed weight. Off-gas from pretreating contains tars, tar acids, carbon oxides, sulfur dioxide, water vapor, and sometimes small amounts of oxygen.

In the utility gas process, we propose to pretreat coal at gasifier pressure and to comingle pretreater off-gas with gases from the gasifier. Hot pretreater char is fed directly into the gasifier, thus avoiding thermal losses. This design eliminates the waste-water and gas-treatment problems associated with low-pressure pretreatment. Furthermore, a smaller pretreater vessel is needed for pressure operation. The overall plant complexity is reduced and so is its capital cost.

The heat of reaction in the pretreater is removed by generating steam in heat exchanger tubes contained in the fluidized bed. The amount of steam generated is more than the gasifier steam requirement.

THE GASIFIER

We want to obtain rapid gasification rates which will permit a higher throughput for a given reactor size and, therefore, will result in a less costly plant. Rapid, precise control of the gasifier operation is needed to follow changes in the power demand. Production of a clean, low-carbon ash is a primary economic consideration.

The gasifier is designed to gasify coal with air and steam in a fluidized bed. Simultaneously, the coal ash is agglomerated into larger and heavier particles for selective separation from the bed. The principle of ash agglomeration and separation was discovered by A. Godel[1] and developed into the Ignifluid boiler. The concept was described by Jequier et al.[2,3] following laboratory and pilot plant gasifier development at Centre d'Etudes et Recherches des Charbonnages de France. We have adapted and modified the Jequier design in development of this reactor concept.

We call the gasifier the ABR (Agglomerating Bed Reactor). The ABR concept resolves the main disadvantage of coal gasification in a fluidized bed rich in carbon: How can low-carbon-content ash be selectively removed from the bed? Advantages of fluid-bed gasification are retained. These are:

1. Bed temperature can be uniformly and readily controlled.
2. High reaction rates can be attained because of excellent gas-solids contact and large surface area of the solids.
3. Coal fines from mining and crushing can be used in the feed.
4. The mass of carbon in the fluid bed ensures reducing conditions at all times.

The ABR is fluidized by a mixture of air and steam. Gasification takes place at about 1900°F in the fluidized bed. Part of the fluidizing gas enters through a grid which is sloped toward one or more cones contained in the grid. Heavier particles migrate along the sloped grid toward the cones. The rest of the fluidizing gas flows upward at high velocity through the throat at the cone apex, creating a submerged jet within the cone. The temperatures generated within the jet are somewhat greater than in the rest of the bed. As carbon is gasified in and near the jet, ash is heated to its softening point. The sticky ash surfaces cling to one another, and ash agglomerates grow in the violently agitated jet. When heavy enough, the agglomerates fall counter to the high-velocity gas in the throat and are thus separated from the fluid bed.

To protect the ash lock hoppers from the hot agglomerates, they are filled with water which is boiling from the heat contained in the ash. The steam generated reduces the amount of external steam needed for the ABR. When filled, the hopper is flushed into filters to recover a wet cake of ash for final disposal. Filtered water returns to the lock hoppers.

TAR AND DUST REMOVAL

Above the ABR fluid bed we have designed for a gas residence time of 10 to 15 seconds; the gas temperature will be between 1500 and 1900°F. By "soaking" the gas at high temperature, tars and oils which may be evolved are thermally cracked to gas and carbon. Elimination or reduction of tars and oils in the raw gas will reduce heat exchanger fouling and will simplify by-product and waste-stream cleanup and treatment.

occur in the expander. Having defined the gas condition at the expander exhaust, the desired moisture content of the gas is obtained by adjustment of the main gas separator temperature. With a 300-psia gasifier, this temperature is 224°F; with a 1000-psia gasifier, it is 300°F. The gas expander inlet temperature is adjusted to give the desired exhaust temperature. Power recovered by gas expansion is used to drive the gasifier air compressor. The air compressor power requirement is about 19 bhp/ton-day of coal feed in a 300-psi plant and 26 bhp/ton-day in a 1000-psi plant.

Condensed water from the main gas separator and from the lock hopper gas separator is fed into a low-pressure stripping tower to remove dissolved gas. The stripped gas rejoins the main gas flow entering the Stretford scrubbing tower. Stripped waste water is cooled and sent to either biological treatment or active carbon treatment for phenol removal.

The advantages of the proposed design are: (1) the gasifier air compressor is driven by process energy, (2) the main gas stream is not water-cooled to obtain the desired 120°F, (3) a minimum of waste water is produced, and (4) the conditions of heat recovery to the power cycle are well defined so that an efficient recovery system can be designed.

COOLING WATER AND WASTE TREATMENT

Cooling water requirements are minimal in this utility gas plant design as a result of some of the process choices that were made. These are: (1) use of a dry feed system, (2) pretreatment at pressure, (3) gas expander exhausting at 120°F, (4) gasifiying at higher pressure, and (5) use of the Stretford process for sulfur removal. In addition, air cooling is used where feasible.

Waste-stream treatment problems associated with coal gasification plants can be serious. We reduced the severity of these problems by choosing the dry feed system, pressure pretreatment, single-stage high-temperature gasification with ash agglomeration, raw gas "soaking" at high temperature, and the Stretford process.

SYSTEM PERFORMANCE

Table 1 shows the calculated plant performance for 300-psi and 1000-psi utility gasification plants. Note that although more methane is formed in the 1000-psi gasifier, the product gas heating value is not greatly different for the two cases. Higher-heating-value gas can be produced if the gasifier is designed as a two- or three-stage unit with gasification temperatures increasing progressively in each stage. We do not believe that this increased complication is warranted. Also, higher-heating-value gas can be made if the pretreating unit off-gas is diverted from the gasifier. This will involve additional plant equipment for condensing, separating, and waste-stream treating of the off-gas and its components. This, too, is considered to be an undesirable plant addition.

Table 1. CALCULATED PLANT PERFORMANCE

Gasifier pressure, psia	300		1000	
Product gas	Wet	Dry	Wet	Dry
Heating value, Btu/scf	140	153	150	164
Composition, vol %				
CO	17.8	19.4	12.5	13.7
CO_2	9.2	10.0	13.6	14.8
H_2	12.1	13.3	11.6	12.6
H_2O	8.5	..	8.5	..
CH_4	4.3	4.7	7.1	7.8
N_2	48.1	52.6	46.7	51.1
Total	100.0	100.0	100.0	100.0
Thermal efficiency, %				
To all products	86.5		88.2	
To gas only	73.2		73.9	
To steam only	12.7		12.6	

The thermal efficiency of the plant is very good in both cases, with the 1000-psi plant being slightly higher. The thermal efficiency is determined by comparing product heating values, heat contained in net steam, etc., to the heating value of the coal feed. Compare the 86 percent efficiency of the utility gas process with the 66 percent efficiency of coal gasification plants producing synthetic pipeline gas.

The gasification plant output should respond at nearly the same rate as the power plant responds to electrical demand changes. The ABR gasifier design should be operated nearly all of the time; its temperature should not fluctuate drastically or serious internal refractory damage may result. However, the throughput can be cut back significantly, and in this way the plant may serve as a load-following plant in addition to its use in base-load operation. If the plant is initially operating at design capacity, the first move in decreasing output is to decrease steam and air flow to the gasifier. This flow change can reduce plant output by a factor of 3 or 4 very rapidly. If a further reduction is needed, the ratio of air to steam is reduced, causing a slow decline in gasifier temperature. By lowering this temperature from 1900 to 1300 or 1400°F, an additional reduction in output of 10 to 20 is obtained. The gasifier temperature should be held within a few hundred degrees of its normal operating temperature most of the time to avoid thermal shock and consequent cracking and spalling of the gasifier internal refractory. Therefore, if the boiler plant is shut down temporarily, we prefer that the gasifier operation continue at minimum rates and that the produced gas is burned in a flare or burning pit.

CONCLUSIONS

We have described the preliminary design of a coal gasification plant for manufacturing clean utility fuel gas for the electric power industry. Its operation will demonstrate the economics and reliability of such a plant when used to fuel an existing boiler. Modifications in this design will adapt the gasifier concept to combined-cycle power generation and to the manufacture of clean fuel for other industrial uses.

We expect that the plant cost and the product energy cost will be less by a significant amount than those costs for an equivalent-size synthetic pipeline gas plant. Proof of this design will provide electric utilities with a realistic method for conversion of coal to a clean fuel.

REFERENCES

1. Godel, A. A New Combustion Technique. Eng. Boiler House Rev. 71:145-153, May 1956.
2. Jequier, L., L. Longchambon, and G. Van de Putte. The Gasification of Coal Fines. J. Inst. Fuel. 33:584-591, 1960.
3. Jequier, L. et al. Apparatus for Dense-Phase Fluidisation, U.S. Patent 2,906,608. September 29, 1959.

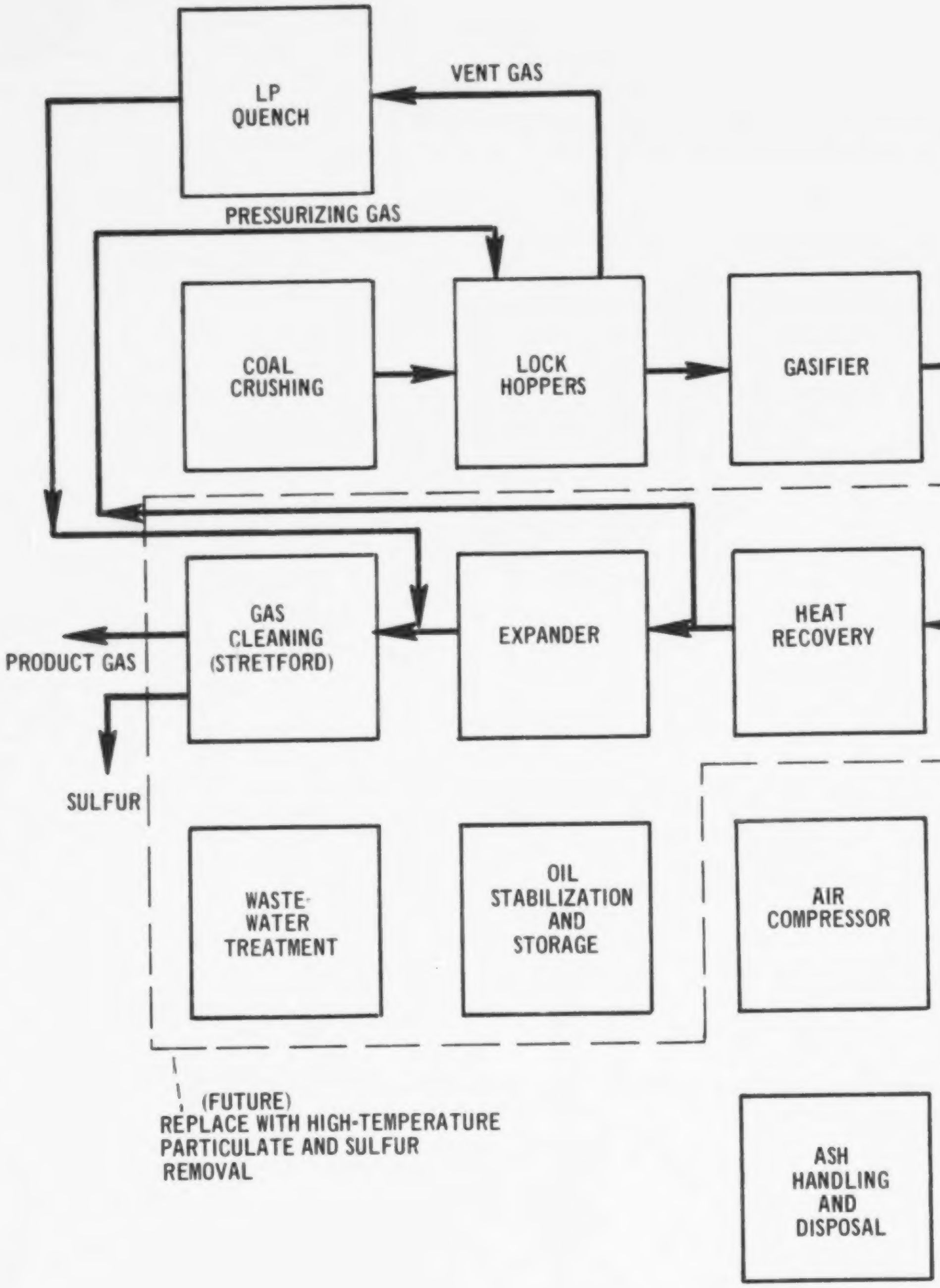

Figure 1. Coal gasification pilot plant block flow diagram for clean utility gas.

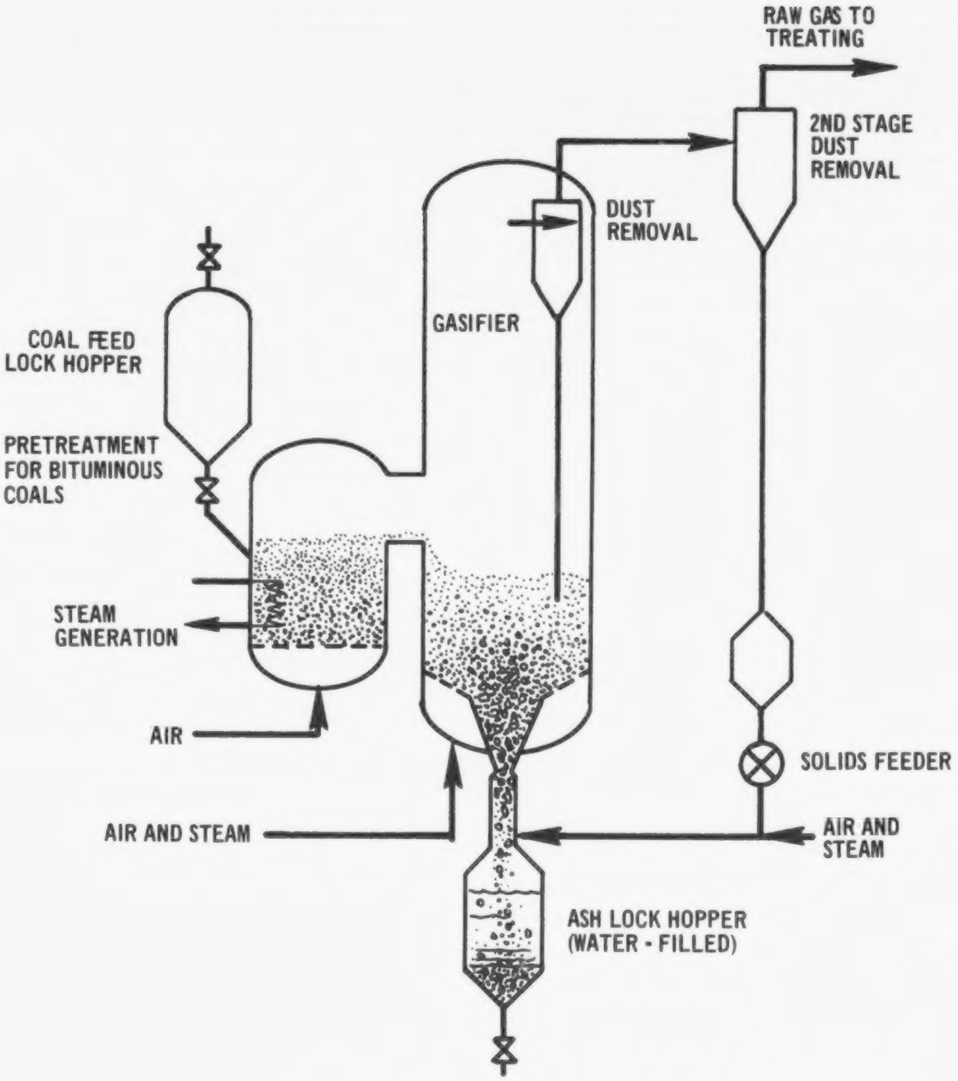

Figure 2. Agglomerating bed reactor.

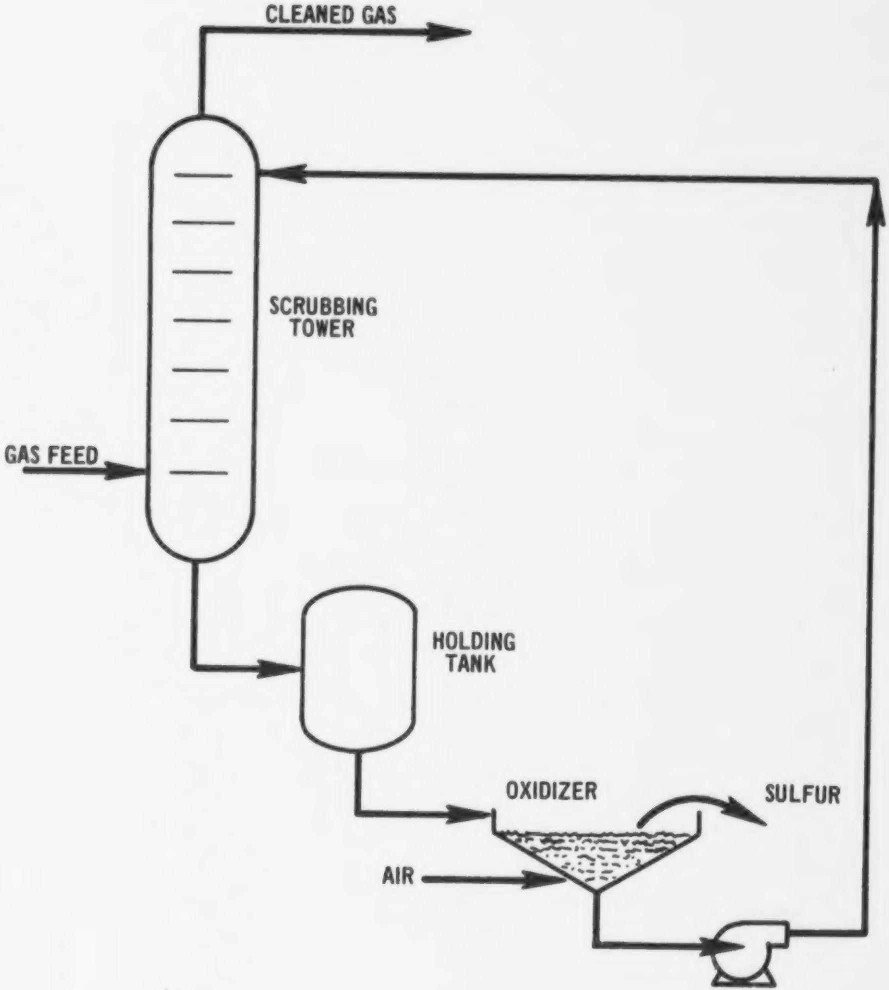

Figure 3. Stretford process.

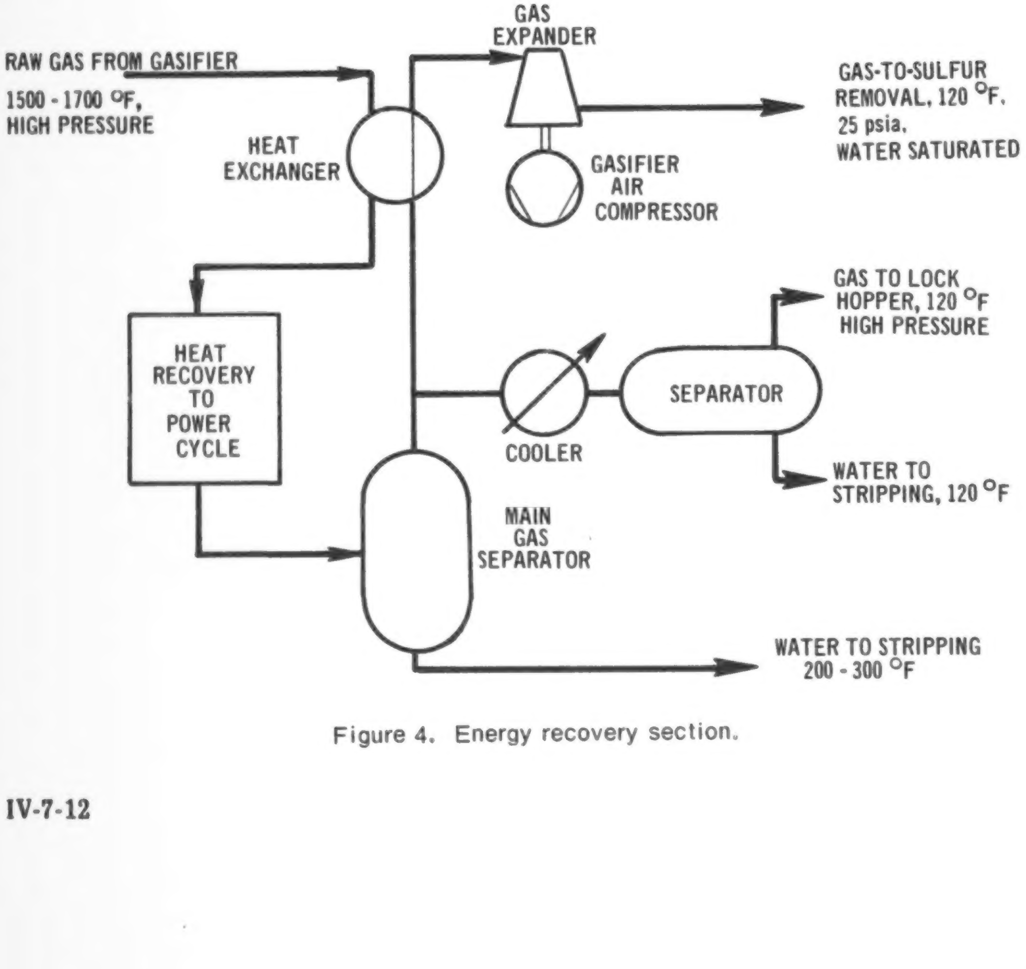
GAS EXPANDER
RAW GAS FROM GASIFIER
1500 - 1700 °F, HIGH PRESSURE
GAS-TO-SULFUR REMOVAL, 120 °F, 25 psia, WATER SATURATED
HEAT EXCHANGER
GASIFIER AIR COMPRESSOR
GAS TO LOCK HOPPER, 120 °F HIGH PRESSURE
HEAT RECOVERY TO POWER CYCLE
SEPARATOR
COOLER
WATER TO STRIPPING, 120 °F
MAIN GAS SEPARATOR
WATER TO STRIPPING 200 - 300 °F

Figure 4. Energy recovery section.

SESSION V:

Pilot Plant Design, Construction, and Operation

SESSION CHAIRMAN:

Mr. H.B. Locke, National Research Development

1. THE DESIGN, CONSTRUCTION, AND OPERATION OF THE ABINGDON FLUIDISED BED GASIFER

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ABSTRACT

A detailed description is given of the design and construction of the desulphurising fluidised-bed gasifier which was built and operated at the Esso Research Centre at Abingdon under the terms of OAP Contract CPA 70-46.

The unit was operated under gasifying conditions for a total of 450 hours during the commissioning period. Information is presented concerning the operational problems which were encountered and the remedial steps which were taken.

INTRODUCTION

The information in this paper supplements that given in reference 1, which provides process data relating to the retention of sulphur in fluidised beds of lime during the *in situ* partial combustion of fuel oil. The Abingdon gasifier is the first of its kind and incorporates a number of unique features which were designed specifically for pilot scale operation.

THE DESIGN AND CONSTRUCTION OF THE GASIFIER

Three primary decisions determined the size, configuration, and mode of operation of the gasifier. These were as follows:

1. The internal configuration of the regenerator of the gasifier was to be identical with that of the batch units which had been used in the exploratory phase.
2. The gas produced was to be burned within a standard packaged boiler fitted with a suitably modified combustion system.
3. A monolithic form of construction was to be used in which all vessels and transfer lines were to be formed as cored holes in a solid block of refractory concrete.

The first decision eliminated one area of uncertainty because it was known from experience that the batch units functioned satisfactorily under regenerating conditions. It also set a limit to the capacity of the unit based on what was then known concerning the capacity of the batch units. At the time that the decision was made these had only been operated at a superficial gas velocity of 4 ft/sec. At an assumed SO_2 concentration of 10 percent by volume in the regenerator off-gas, this gave a sulphur handling capacity of about 8.6 lb/hr. In the case of a 2.2 percent by weight sulphur fuel oil this limited the fuel throughput to 391 lb/hr, giving an energy throughput of 7.1 x 10^6 Btu gross/hr or 6.7 x 10^6 Btu net/hr. The gasification of 391 lb fuel oil/hr at 900°C and a gas velocity of 4 ft/sec with 20 percent of stoichiometric air indicated a cross-sectional area for the gasifier of 4.8 ft^2.

The decision to use a packaged boiler rather than a flare for the second stage of combustion was influenced by a number of considerations. A flare might have had an unfortunate impact on local public relations; it was in any case considered desirable to use the gasifier to fire a standard piece of equipment in order to demonstrate it as a practical proposition. The capacity of the boiler which was available was 10 x 10^6 Btu/hr delivered as pressurised hot water. This appeared to be quite suitable for use as an energy sink for a 7 x 10^6 Btu/hr gasifier; it was decided to dissipate the output to the atmosphere via a pressurised heat exchanger and an atmospheric evaporative cooler. What was not realised at the time, was that it was possible to operate the batch units at up to 8 ft/sec superficial gas velocity. Consequently, although on the original design basis the heat disposal equipment provided a reasonable margin of spare capacity, it subsequently turned out that heat disposal was a factor restricting the range of operating conditions which could be explored.

The use of the packaged boiler enabled flue gas recycle to be used to control the gasifier temperature to levels lower than those dictated by adiabatic operating conditions. This was advantageous since steam would most likely have been used for this purpose in other circumstances, involving the use of an additional utility.

So far as the gasifier was concerned, the choice of monolithic construction imposed its own logic upon the geometrical configuration. The reasons for choosing monolithic construction were: (1) the absence of joints between the various components enabled them to be grouped in a very compact arrangement; (2) the position of the bed transfer ducts in the heart of the block enabled the sensible heat of the transferred bed material to be conserved to the same degree as it would be in a large scale unit; and (3) this was a very simple and cheap form of construction.

The major disadvantage with monolithic construction is that it does not readily lend itself to modification. It was necessary to be sure that transfer lines would work satisfactorily before they were cast in refractory concrete. An additional disadvantage was the reliance on the durability of the concrete when subjected to temperature stresses. In fact some minor cracks did develop but these were found to be self-sealing under gasifying conditions.

When the choice of monolithic construction was made it was found expedient to adopt a rectangular cross section, because this enabled the casing to be constructed from flat, edge-stiffened panels. It was then found that the overall height of the unit was dependent on the size of the gasifier cyclone; the decision was finally made to use two smaller cyclones instead of one large one. This gave the configuration shown in Figure 1. It can be seen that the gasifier bed is rectangular in plan and that the regenerator is between the two gasifier cyclones to one side of the gasifying reactor.

The bed transfer system which was selected was an adaptation of a system proven in commercial practice, where it is employed to control the rate at which bed material descends through a series of stacked beds. The adaptation was necessary in order to enable the system to be utilised to transfer bed material between two beds in parallel and at the same height. In this system the bed transfer duct is almost vertical, but incorporates a horizontal section at its lower end. In the absence of any external agency the duct simply fills with static bed material. When a pulse of gas is introduced into the horizontal section, however, bed material flows down the duct under the influence of gravity. The adaption involved utilising the two fluidised beds as lift pumps for the bed material, each bed discharging the material into a cavity at the top of a transfer duct. The two transfer ducts can be seen in dotted outline in Figure 1 and they are situated on either side of the regenerator in plan. The existence of these transfer ducts adjacent to each of the cyclones suggested that they might also be used as cyclone drains. This was in fact done and in operation half of the gasifier cyclone fines are returned to the gasifier itself and half are passed on to the regenerator. The fines leaving the regenerator bed are trapped by an external cyclone and drained from the system.

It was necessary to test a novel bed transfer system of this complexity before casting it in concrete. This was done by building the full scale cold rig illustrated in Figure 2. The bed material used in this rig was a crushed brick of roughly the same density and size distribution as the lime. Although the gas velocities were matched in the fluidised beds, there was a strong element of conservatism in the operation of the transfer system; no allowance was made for the very considerable volumetric expansion of injected gas under hot conditions. It was also anticipated that the higher gas viscosity at the normal operating temperature would improve the flow properties of the bed material. Tests run with this rig soon showed that the introduction of the cyclone fines into the transfer ducts via simple branches led to severe bridging problems. It was deduced that this bridging was caused by fines being blown back up the transfer lines to block the interstices between the particles of descending bed material.

A way out of this difficulty was found by devising the mixing cavity shown in Figure 3. The shape of this cavity is such that a pocket of gas is trapped within it, leaving a free surface of bed material above the horizontal section of the transfer duct. The theory behind this design is that when the transfer duct is activated, bubbles of gas rise into the pocket and displace gas already there; this gas reenters the free surface of bed material and proceeds up the transfer duct. In this way the free surface can act as a filter for fines. In practice the device which was built of plexiglass worked very well. Lenses of fines were trapped by the coarser solids, and these inclusions moved down towards the horizontal transfer duct where they disappeared. This modification solved the bridging problem.

Other design problems related to the thermal expansion of the refractory block when it was brought up to operating temperature. There was also the question of thermal insulation to be dealt with. The construction of the unit allowed the transverse thermal expansion in a very simple fashion. The plates forming the casing of the gasifier were lined with 3 in. thick slabs of 50 lb/ft^3 castable insulating material. When the casing was assembled the inner surface was lined with 1/4 in. thick expanded polystyrene sheet; the refractory concrete block was cast within this lining. The expanded polystyrene was subsequently melted out leaving a gap between the refractory block and the insulation. The vertical expansion of the block posed more difficult problems because it was necessary to connect the gasifier to the burner in a gas-tight manner, but which allowed 3/8-in. relative vertical movement. Since there were two gasifier outlets it was necessary to employ a Y-shaped bifurcated duct consisting of a mild steel casing enclosing a refractory concrete lining in which the gas passages are cast (Figure 4). A layer of calcium silicate slab insulation is sandwiched between the refractory concrete and the steel casing to minimise heat losses.

The duct is suspended immediately above the cyclone outlets with an expansion allowance between the corresponding faces. The expansion joint is sealed by a stainless steel bellows to provide a gas-tight assembly. The duct is supported at the other end by a roller which accommodates the horizontal expansion of the duct and burner; it is sealed to the boiler face with a compressible insulating seal.

The burner design presented some problems because there was no information available concerning the combustion characteristics of this hot gas. Some simple burners had been tested on the early batch units which showed that the gas would burn easily. By introducing some premix air to the burner it was possible to burn the gas with a steady smoke free flame and low excess air.

The 7 x 10^6 Btu/hr burner used for the continuous gasifier is illustrated in Figure 5. It consists of two sections--a premix zone in which about 10 percent of the combustion air may be introduced and a main section in which the balance of the air is added. In both of these sections an inner stainless steel assembly is used which is insulated from the cold combustion air introduced around the assembly. The insulation is necessary to maintain the temperature of the hot gas duct which otherwise might become obstructed with condensing material from the hot gas.

These inner insulated assemblies are fixed to the outer casing at one end and are free to expand along the axis of the burner at operating temperature. The gas issues from the burner to mix with the main combustion air through a stainless steel orifice sized to give a pressure drop between 3 and 4-in. water gauge.

For safety reasons it was decided to use a continuous pilot flame on the experimental plant; here problems arose because this flame must be stable at normal running conditions which means a high gas and air rate, unlike a conventional burner which lights off a pilot at a low flame setting. The problem was overcome by placing a small stainless steel deflector plate to shield the flame of the pilot from the main air.

The only other problem with the burner arose from gas turbulence at the entry to the burner orifice nozzle which threw out deposits in the burner inner duct. This was overcome by smoothing the gas flow into the orifice by a suitable entry duct; no further deposits were observed.

Because the overall height of the gasifier is considerably greater than the centre height of the boiler furnace, it was necessary to place the gasifier in a pit in order to line up the two components. A general plant layout is shown in Figure 6.

The air distributor of the gasifier is provided with horizontal nozzles made by drilling six radial holes of 0.177-in. diameter through each of 32 stainless steel capped tubes. The distributor and its plenum form a removable box structure built of mild steel; the top of the box, from which the nozzle assemblies project, is covered with refractory concrete.

There are a number of penetrations through the walls of the gasifier to provide access for thermocouples, manometer probes, fuel injection tubes, bed drains and the gas injectors used to activate the bed transfer system. These penetrations were all cored before the block was cast. In addition, a precast quarl for the startup burner was also placed in position before the refractory concrete was cast.

Figure 7 shows the lower core assembly of the gasifier during an early stage of construction. The cores for the transfer system were made of plexiglass and were filled with wax in order to avoid the possibility of damage and filling with concrete during the casting operation. Figure 8 shows the unit after the first two lifts had been cast.

G.R. Stein Refractories Ltd. advised on the method of construction and built the unit using their refractory concrete Durax C.1600. This material contains about 50 percent alumina, 42 percent SiO_2, 5 percent CaO, with traces of Fe_2O_3 and MgO. The maximum operating temperature is 1600°C with a melting point of 1710°C.

In the first instance the only internal metal components of the gasifier were the cyclone outlet tubes. These proved to be unsatisfactory and were subsequently replaced by tubes of self bonded silicon carbide.

THE ASSOCIATED SYSTEMS

The pilot plant flow plan is shown in Figure 9. It will be seen that the startup burner is fired by propane. This burner is used to heat the gasifier to its working temperature and is a standard commercial burner with a variable output, delivering 700,000 Btu/hr at the maximum firing rate. Propane is also used to fuel the pilot burner fitted to the boiler. The main fuel system utilises three small metering pumps which draw fuel oil from a circulating stream in a ring main and deliver it to the three fuel injectors of the gasifier. A small amount of air is injected with the oil in order to prevent coking in the injector tubes. A switch from fuel oil to kerosine is also provided. This enables the consumption of propane during the warm-up period to be reduced while avoiding the introduction of sulphur into the gasifier. The flue gas recycle system which is used to control the temperature of the gasifier is also shown in the flow plan. The flue gas recycle stream is first cleaned in a cyclone, then passed through an orifice plate flowmeter and a control valve to the inlet side of the first gasifier blower. A second control valve throttles the air supplied to this blower; by making suitable adjustments to these two valves it is possible to vary both the total supply of gas to the plenum of the gasifier and the composition of the gas in terms of the proportions of flue gas and air which it contains.

The operating temperature of the regenerator tends to be self regulating when no oxygen is present in the tail gas, because CaS can yield two oxidation products — $CaSO_4$ and $CaO + SO_2$. The first reaction releases much more heat than the second reaction; but as the temperature rises the second reaction tends to predominate so that in effect the calorific value of the sulphur fed to the regenerator tends to fall. In order to hold the temperature at a specified level however the bed transfer system is arranged to automatically increase the transfer rate when the temperature falls below the set point. Because of the temperature difference between the two beds, the consequent adjustment to the rate of heat transfer from the regenerator to the gasifier brings the regenerator temperature into line. The rate of SO_2 release at any set regenerator temperature depends on the rate at which air is fed to the regenerator. For experimental purposes this is a manual adjustment; when completely automatic control is used, the air rate to the regenerator may be controlled to hold the O_2 concentration in the regenerator tail gas at a constant level. The return of solids from the regenerator to the gasifier is controlled by the pressure drop across the regenerator bed.

Equipment was not installed to deal with the 10 percent SO_2 stream from the regenerator because this did not form part of the development programme at this stage. The SO_2 stream from the pilot plant was therefore connected into the boiler stack, thus creating a flue gas identical to that resulting from direct combustion of the test fuel.

The instrument flow plan for the installation is shown in Figure 10. This, however, does not show the packaged pressurisation system which maintains constant boiler water pressure and temperature.

Considerable attention has been given to safety measures; the plant is protected by a number of sensor systems which detect both hazards and conditions. Signals from the sensors will, according to a predetermined selection, either shut down the whole plant and operate an alarm, or give an alarm and a visual indication of the trouble, or merely give a visual indication.

During test runs the unit is operated on a 24-hour shift basis, with one professional and two non-professional personnel in each shift team.

OPERATIONAL PROBLEMS

Three operational problems became apparent during the first attempts to run the unit for a prolonged period:

1. The formation of lime/coke deposits in the ducting between the gasifier and the burner.
2. A tendency to form too large a proportion of $CaSO_4$ within the regenerator.
3. Poor containment of bed fines.

The deposits in the gas ducting were formed locally in areas of high turbulence. In the first instance deposits tended to choke the inlet ports of the cyclones which were originally square edged. This problem was greatly alleviated by chamfering these edges to give a smoother flow transition. Two other critical areas were found where the vertical cyclone exit channels intersected the converging horizontal channels within the Y-shaped duct. The design of the ducting has since been modified to provide a smoother gas passage at these elbows.

The deposits laid down in the gas ducting could be removed without shutting down the unit by a controlled burn-out procedure. A more serious form of deposit was, however, found within the regenerator and within the transfer line from the regenerator to the gasifier. The deposits in these areas built up relatively slowly, but could only be removed when the unit was shut down. It was deduced that the cause of these deposits, which contained no carbon, was the excessive formation of $CaSO_4$ within the regenerator.

The oxidation of CaS in the regenerator is analagous to the oxidation of carbon in the gasifier in that two reactions occur and they appear to occur sequentially. In the gasifier the carbon deposited on the lime is first oxidised to CO_2 which is subsequently reduced to a large extent to CO on passing through the bed. Within the regenerator there is a tendency to form $CaSO_4$ near the distributor which subsequently reacts with CaS to form $CaO + SO_2$. It has been observed by Curren, Fink and Gorin[2] that during the course of this reaction a transient liquid is formed which can cement particles together. This is thought to be the mechanism by which deposits were laid down within the regenerator and in the regenerator to gasifier transfer duct. These deposits were largely composed of calcium sulphate; a photograph of the deposit which was removed from the regenerator at the completion of Run 3 is shown in Figure 11. This deposit was wedge shaped and grew from the wall of the regenerator opposite the outlet of the bed transfer duct from the gasifier. As can be seen in Figure 12 the distributor nozzles under the deposit were themselves blocked by coarse bed material; it seems likely that the initial presence of a dead zone induced deposit formation in this area. The design of the regenerator distributor has since been modified and it is now more similar to that of the gasifier.

The formation of deposits in the regenerator is best avoided by reducing the tendency to form $CaSO_4$. This may be done by introducing the bed material from the gasifier into the regenerator at a level well above that of its distributor.

The effect of this change would be to ensure that both the fresh bed material from the gasifier and the incoming air are brought up to the working temperature before they meet. Under these circumstances there should be a greater tendency to form unstable $CaSO_3$ in a single step; less $CaSO_4$ should be available for the liquid phase decomposition. In the case of the experimental unit this has been attempted by lowering the level of the regenerator distributor.

The gasifier cyclones were not found to be very effective in retaining the fines which were produced by attrition and decrepitation. In normal operation they passed solids at a rate amounting to about 2 percent by weight of the fuel used. It is likely that this poor performance was largely due to the rough surface finish of the cyclones, but it was considered that the best way to prevent solids from entering the boiler would be to reduce the amount of fines produced.

Decrepitation occurs during calcination and may be reduced by lowering the bed replacement rate. The batch results indicate that increasing the depth of the bed will not impair desulphurization efficiency. An indication of the importance of bed losses incurred shortly after the entry of stone into the gasifier was the fact that at the end of Run 3 the vanadium content of the bed was three times higher than could be accounted for on the assumption of a uniform bed life.

There were two mechansims which might have contributed to this effect--decrepitation and elutriation during addition. During these runs the bed material was fed into the gasifier through its lid and fell in a stream past the entry of one of the cyclones. It was thought likely that some of the finer material was swept out of the unit before it entered the bed. In order to reduce this possibility the stone feed system was modified; stone now enters the unit through the wall of the gasifier in the vicinity of the bed surface and at a point remote from the cyclone inlets. Attrition in fluidised beds increases rapidly as the superficial gas velocity rises. The highest gas velocities within the bed occur at the distributor nozzles and are required in order to ensure bed stability. The problem here was to combine bed stability with a low nozzle efflux velocity. The adopted solution was to use two stage nozzles in which the kinetic energy imparted to the gas by the pressure drop through the first nozzle was dissipated prior to the low velocity entry of the gas into the bed via the second nozzle.

During the first phase of operation the unit was run for 450 hours under gasifying conditions; the main structure does not appear to have suffered any significant deterioration. A major objective of future work is to reduce the quantity of fines leaving the bed to a level which will enable us to envisage the construction of a full scale unit which would not require hot cyclones. If this could be done there would be a considerable saving on investment and also an improved operability due to the decrease in deposit formation arising from the simplification of the gas ducting. Finally, it seems reasonable to conclude that the operational problems which have so far been encountered do not appear to be unduly severe or intractable.

REFERENCES

1. Craig, J.W.T., G. Moss, J.H. Taylor, and D.E. Tisdall. Sulphur Retention in Fluidised Beds of Lime Under Reducing Conditions. (Presented at 3rd International Conference on Fluidized-Bed Combustion. Hueston Woods. October 29 - November 1, 1972.) (See Session III, Paper 4 this volume.)
2. Curren, G.P., C.E. Fink, and E. Gorin. Phase II Bench-Scale Research on C.S.G. Process, Research and Development No. 16. Consolidation Coal Company, Library, Pa. Prepared for Office of Coal Research, U.S. Department of the Interior, Washington, D.C. under Contract Number 14-01-0001-415. July 1969.

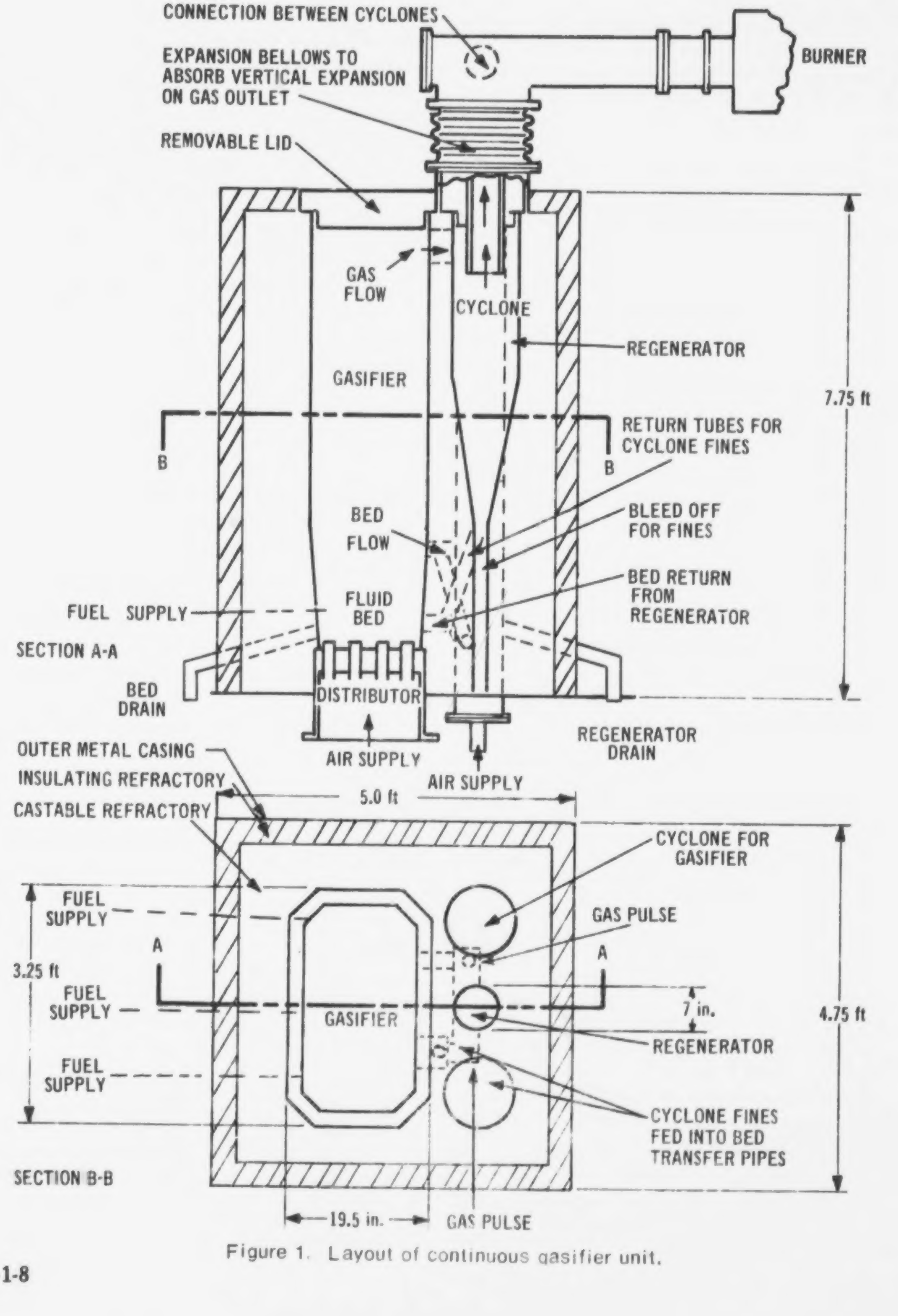


Figure 1. Layout of continuous gasifier unit.

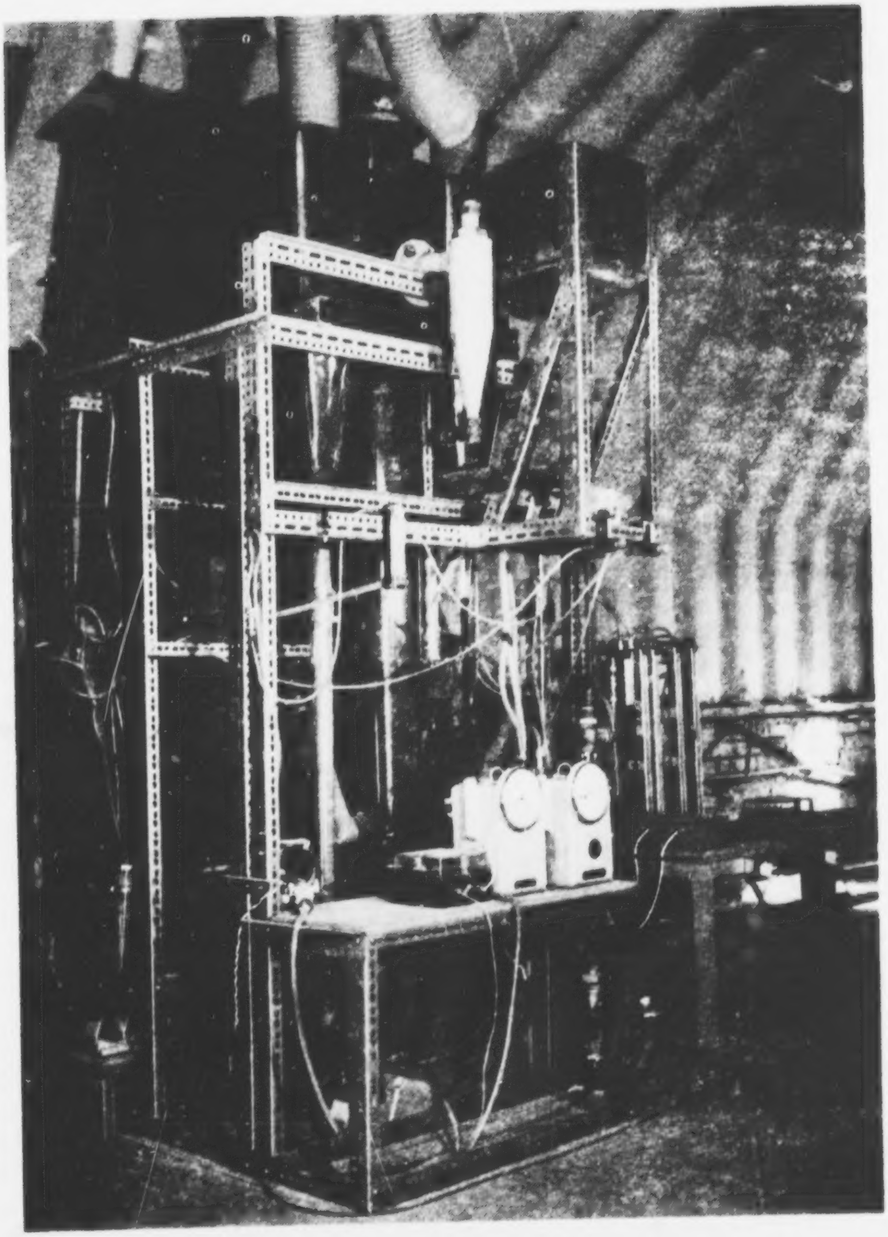


Figure 2. Full-scale cold rig.

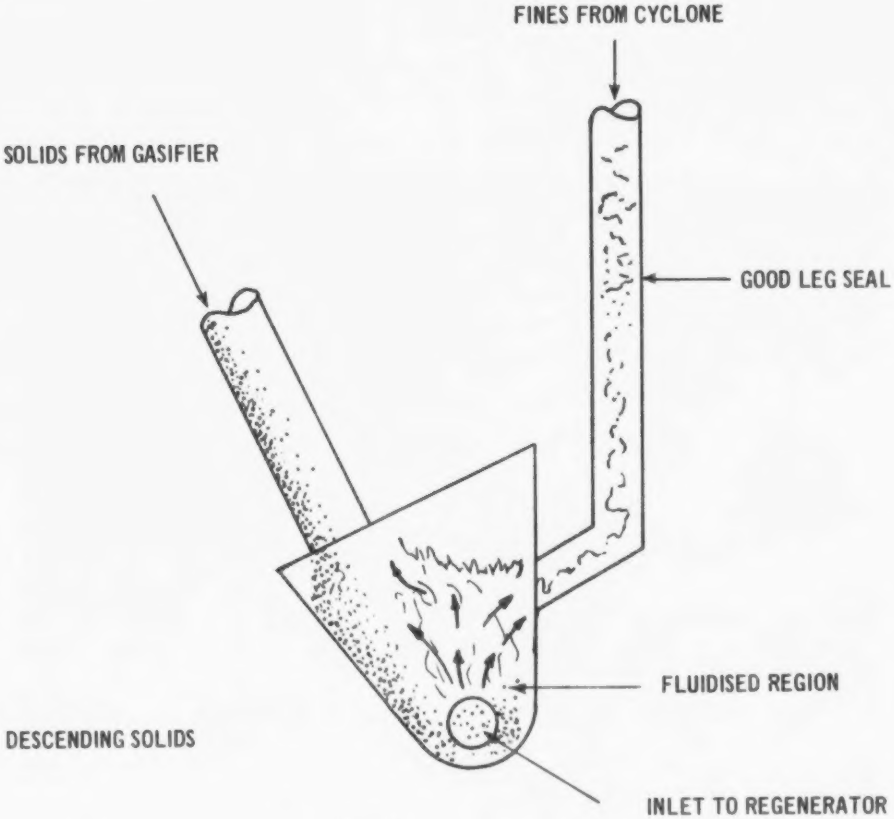


Figure 3. Successful cyclones fines return zone.

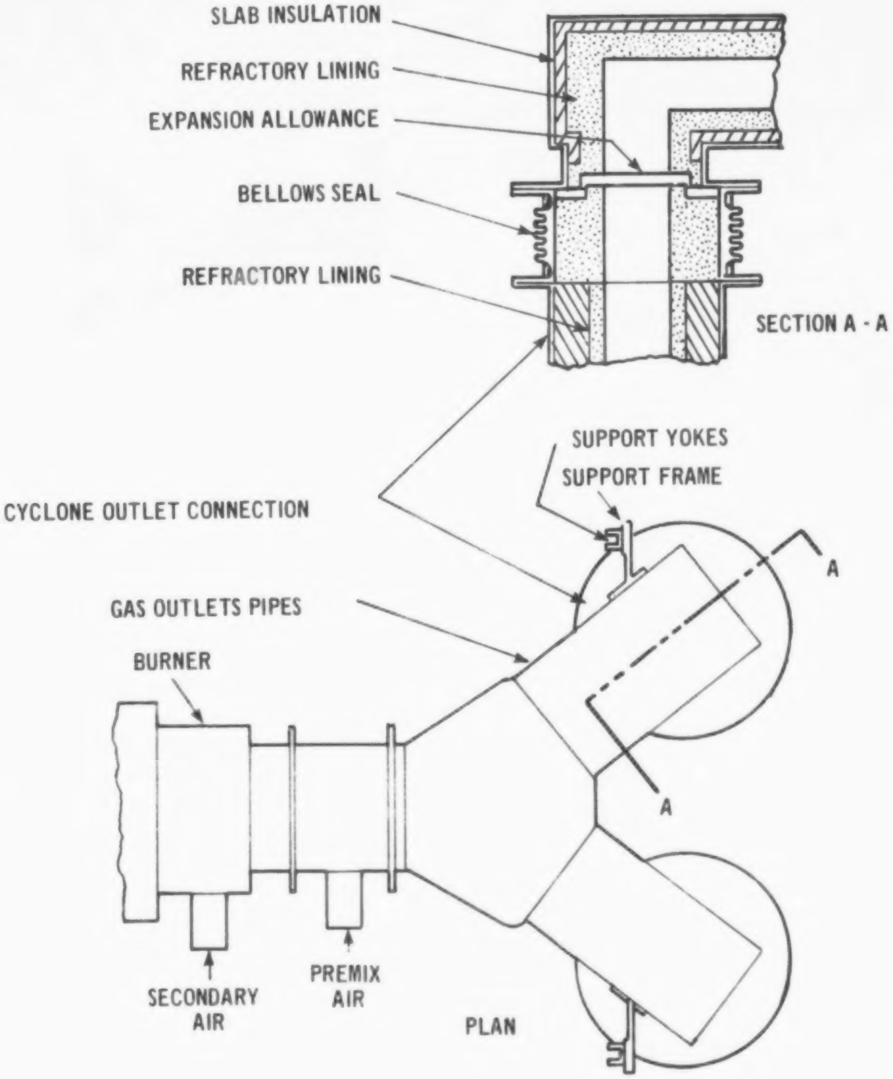


Figure 4. Cyclones to burner manifold.

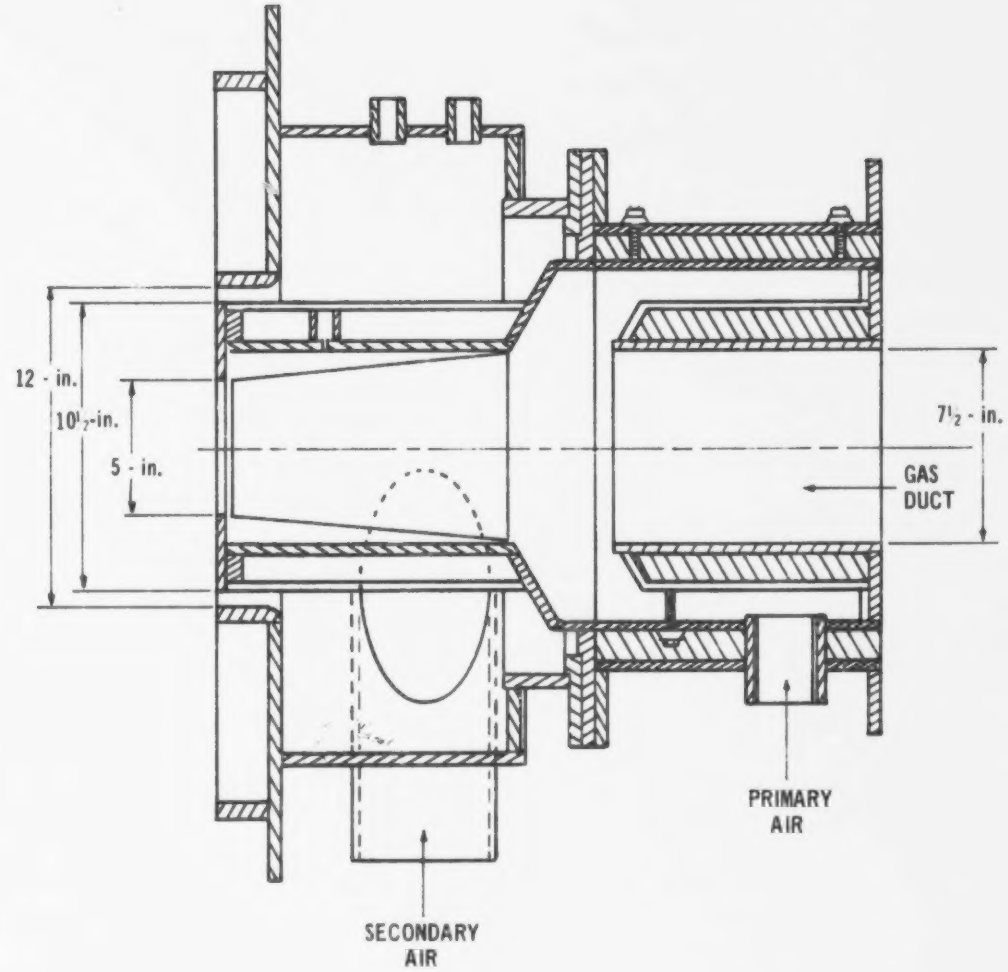
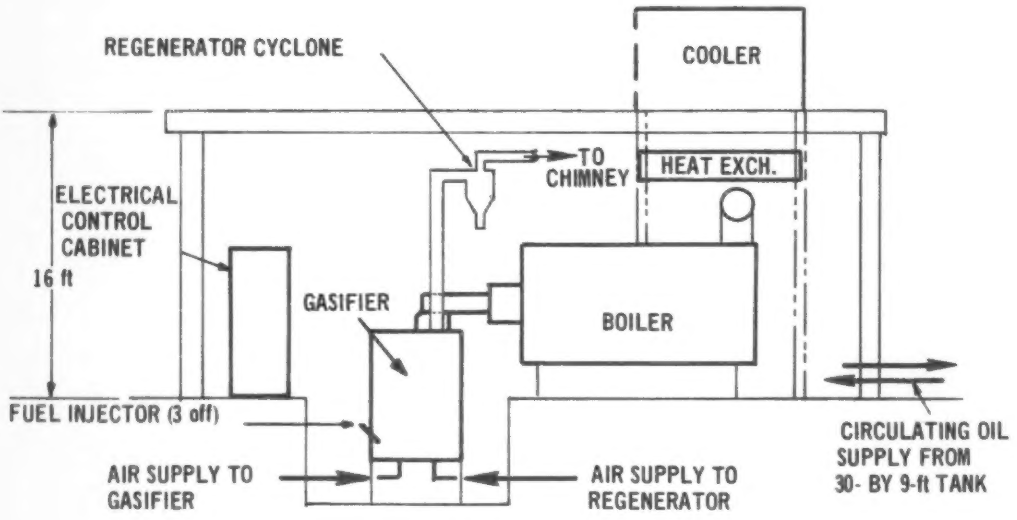
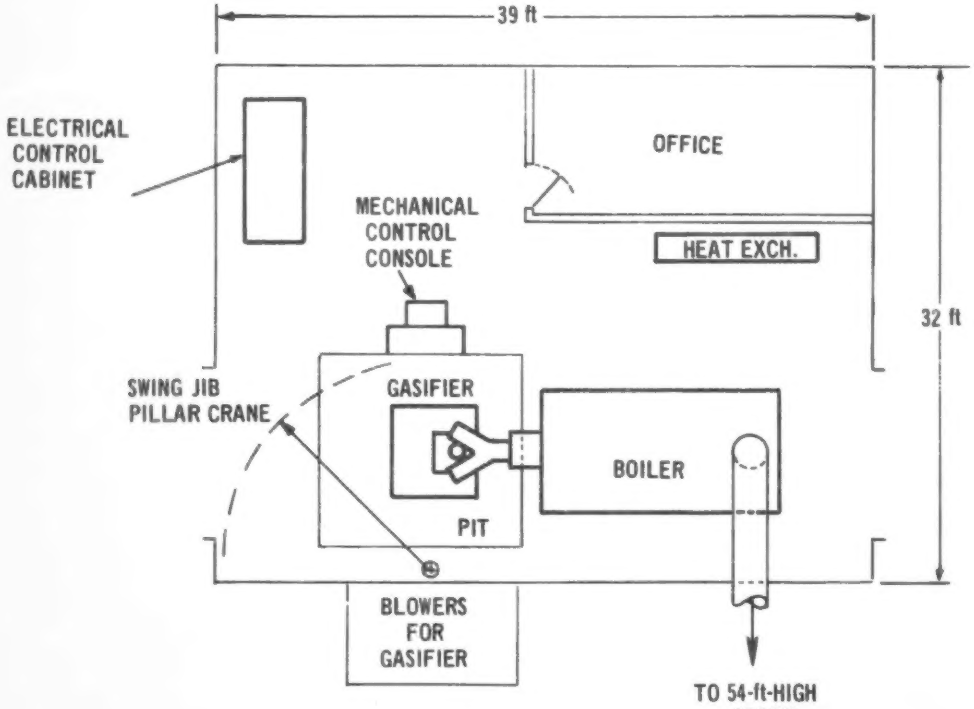


Figure 5. Main gas burner.



SIDE SECTION



PLAN

Figure 6. General plant layout.

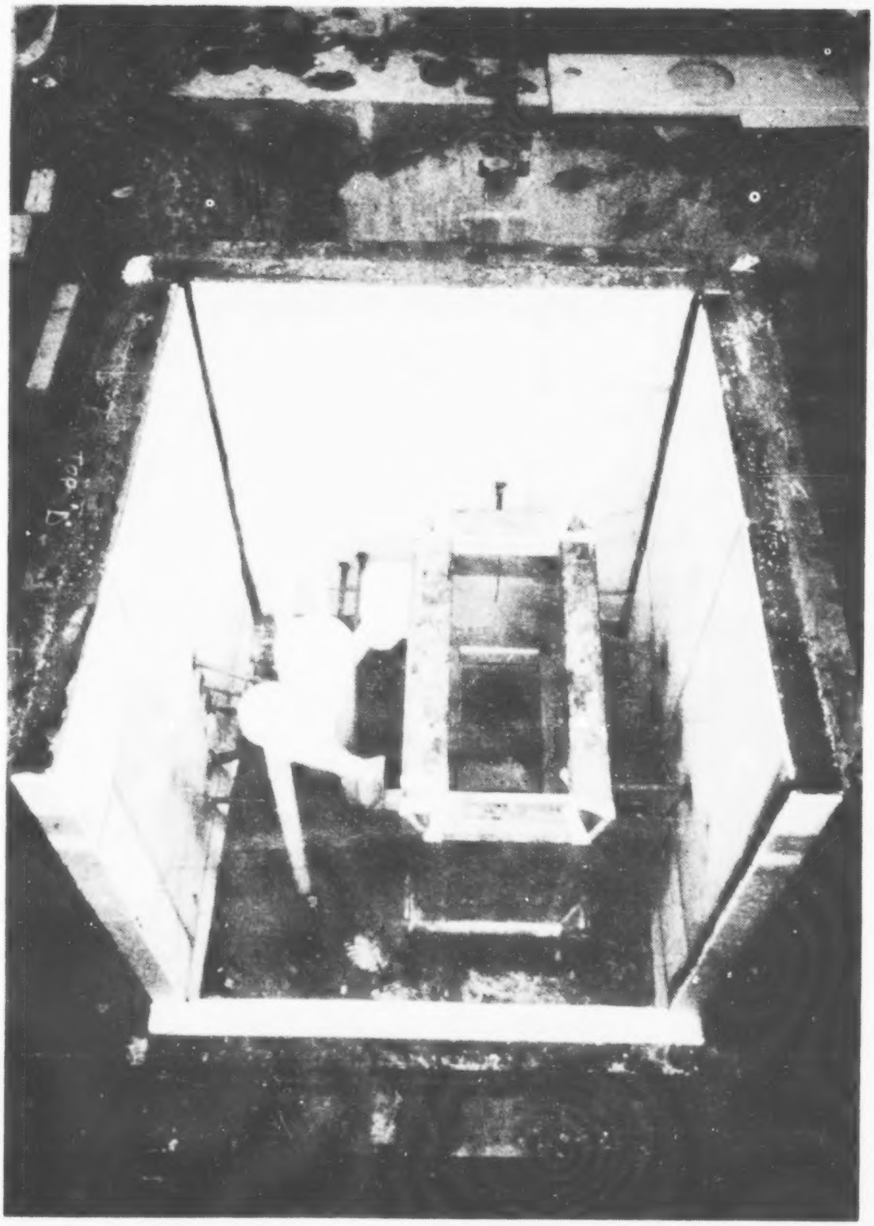


Figure 7. Gasifier lower core assembly during early construction stage.

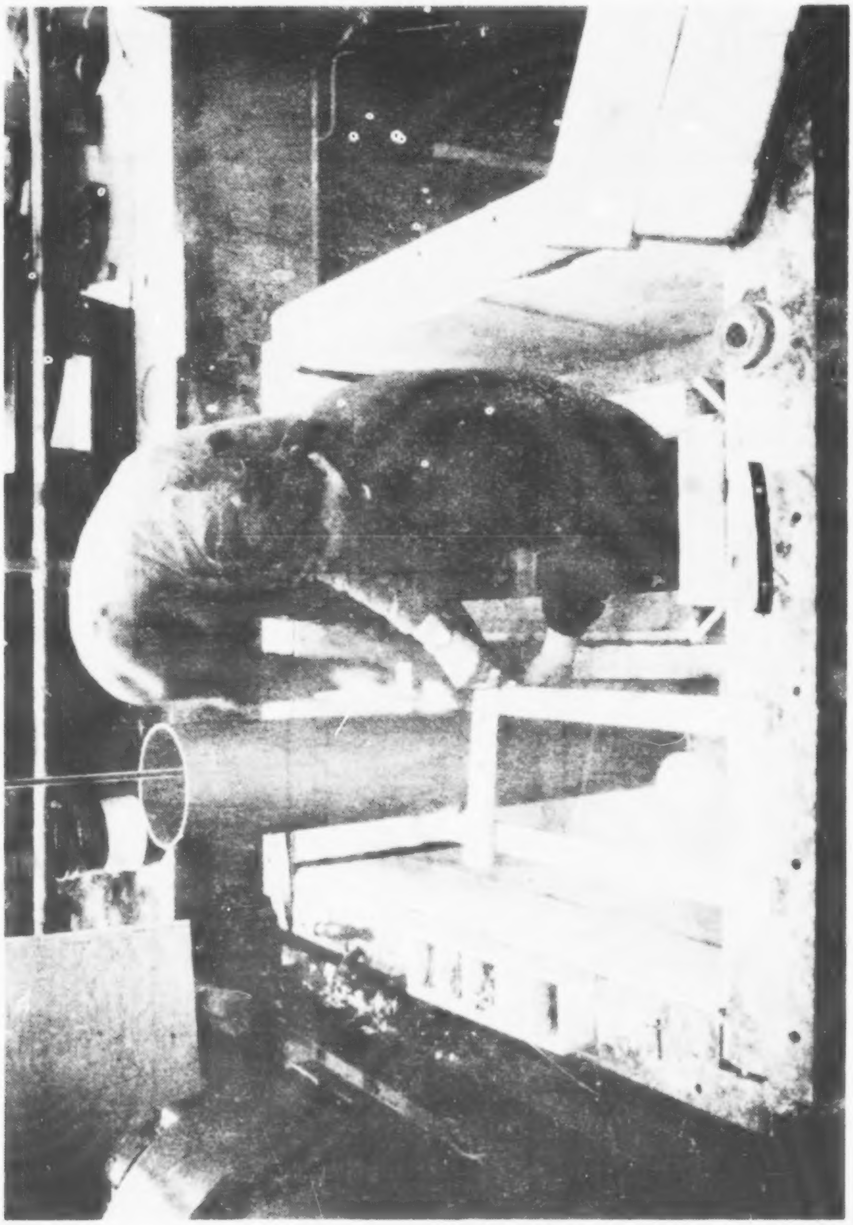


Figure 8. Gasifier lower core assembly following casting of first two lifts.

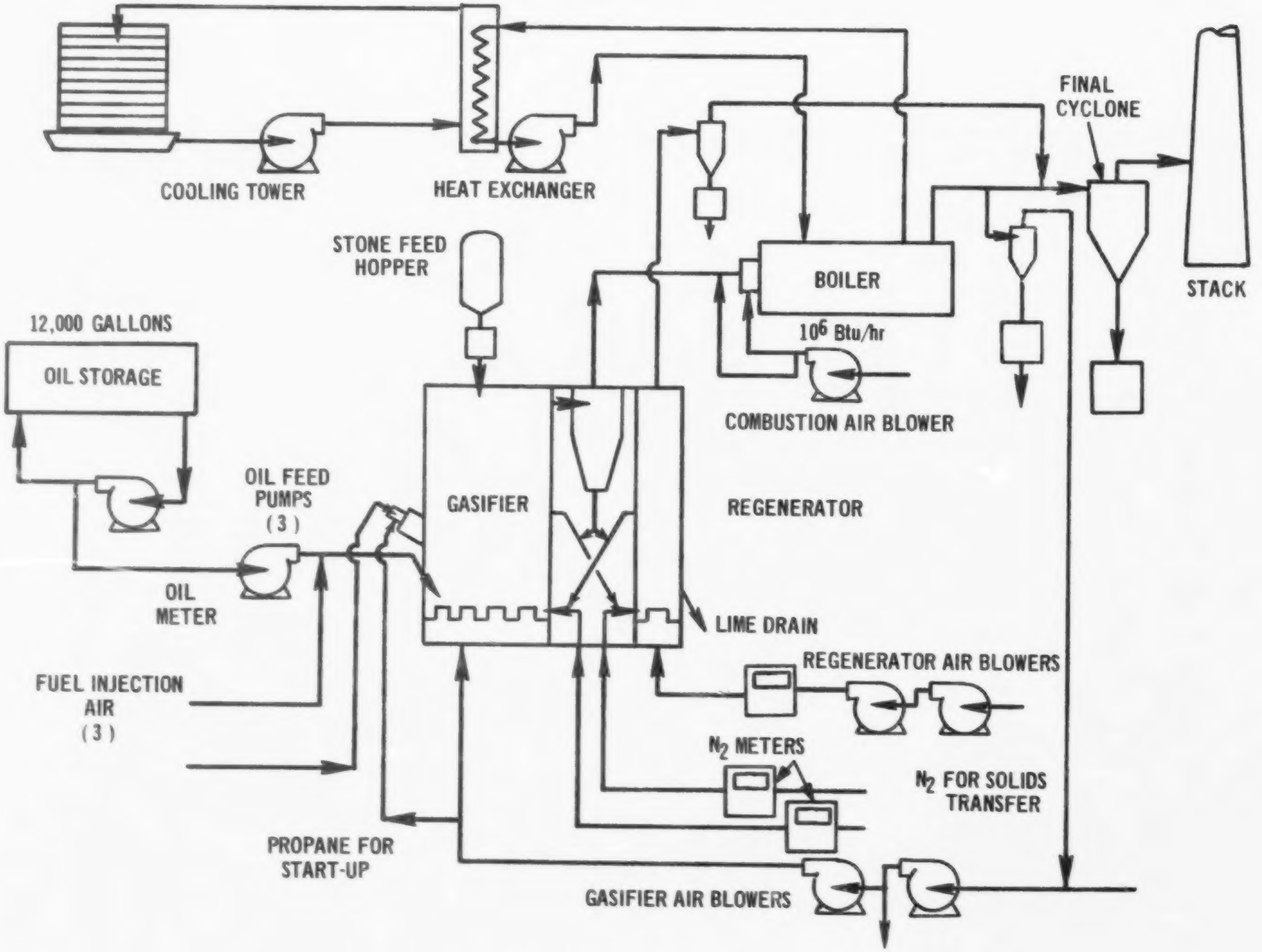


Figure 9. CAFB pilot plant flow plan.

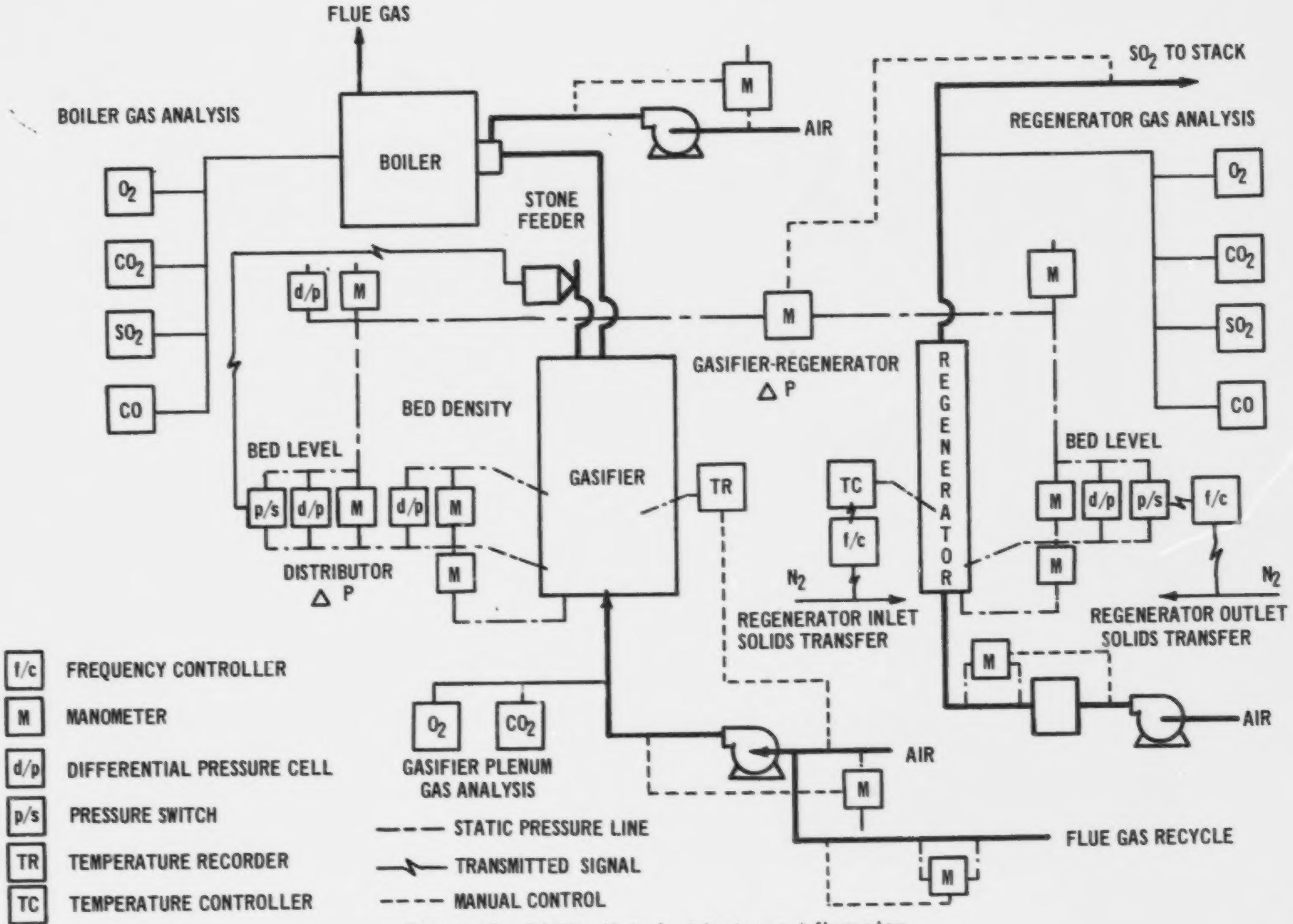


Figure 10. CAFB pilot plant instrument flow plan.

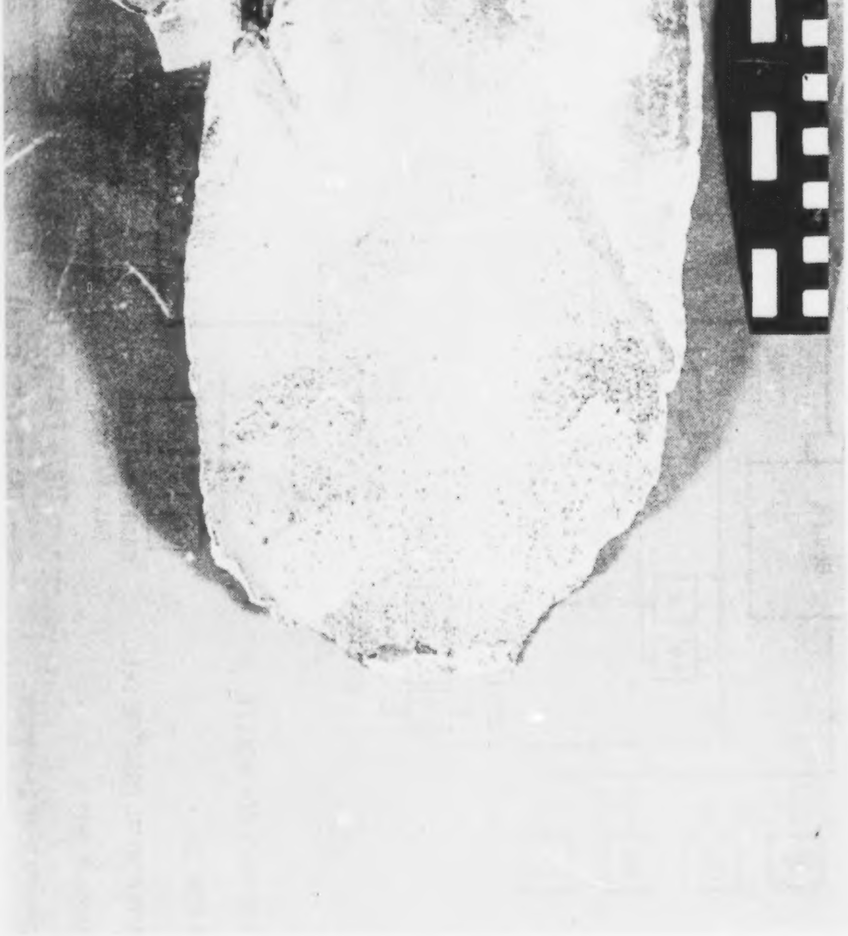


Figure 11. Deposit removed from regenerator after Run 3.

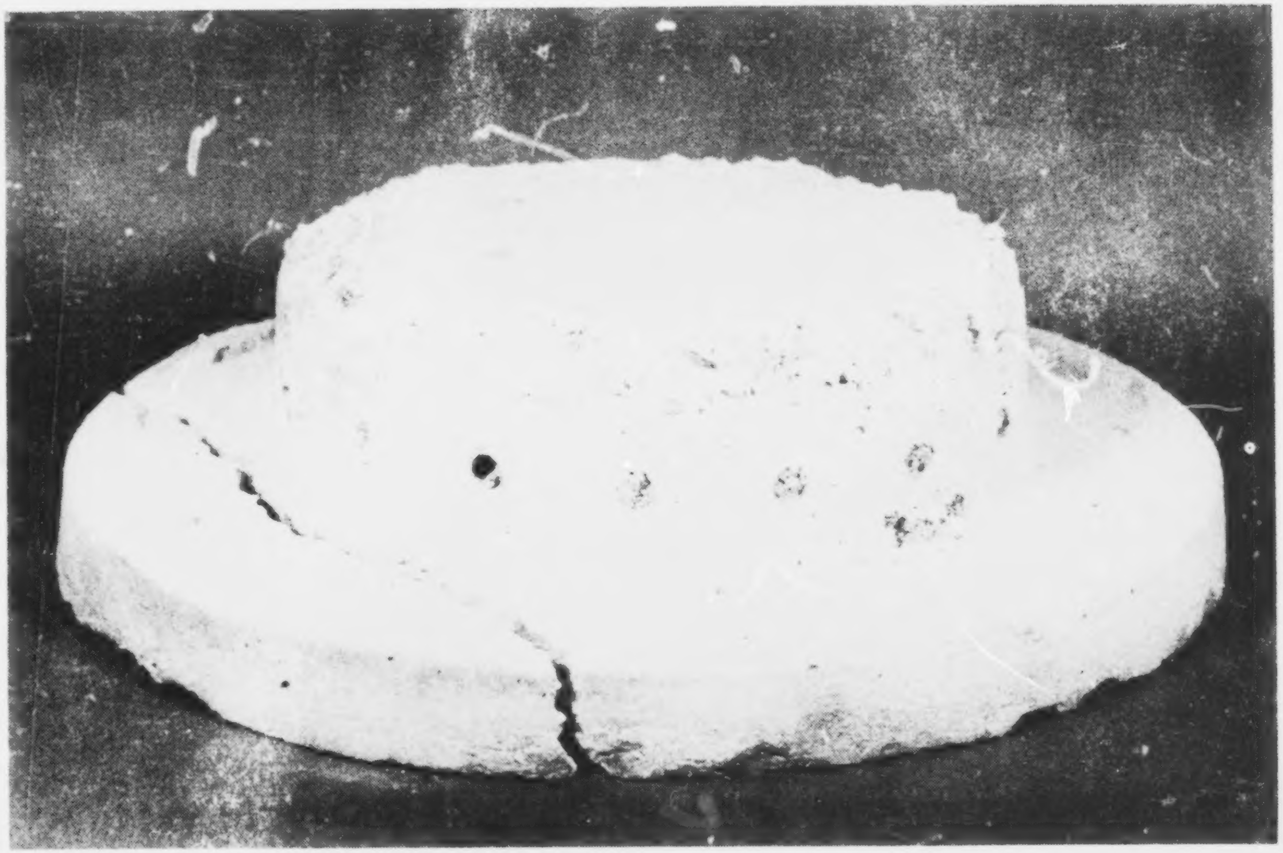


Figure 12. Distributor nozzles blocked by coarse bed material.

2. DESIGN OF FLUIDIZED-BED MINIPLANT

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Esso Research and Engineering

ABSTRACT

Fluidized-bed combustion of coal offers potential both as an efficient compact combustion-boiler system and an air pollution emissions control system. Esso Research and Engineering Company, under contract to the Office of Air Programs of the Environmental Protection Agency, has designed a system capable of fluidized-bed coal combustion and desulfurization with continuous limestone regeneration.

The fluidized-bed miniplant will operate at pressures up to 10 atm and with an input of approximately 6.3×10^6 Btu/hr. This energy input is equivalent to a power plant rated at 635kW (0.63 MW).

In the miniplant, combustion and heat transfer to tubes take place in a reactor containing a fluidized-bed of limestone at 1500-1700°F, providing good heat transfer and an efficient desulfurization reaction between the sulfur dioxide and limestone. The calcium sulfate produced during desulfurization is transferred to an adjacent fluidized-bed reactor and contacted with a reducing gas at 1900-2050°F. This regenerates the lime for reuse in the combustor and produces a by-product off-gas stream of concentrated sulfur dioxide. Thus, regeneration minimizes the limestone feed requirements and the calcium sulfate disposal problems.

The fluidized-bed miniplant design incorporates a 12.5-in. ID combustor and a 5-in. ID regenerator vessel with continuous transfer of solids between these two refractory lined reactors. In the combustor, the fluidizing air enters a plenum, passes through the distributing grid, up through the fluidized bed of solids and the combustion products discharge through two refractory cyclones in series.

Superficial bed velocities and pressures in the combustor and regenerator are automatically controlled. The pressure differential between the two vessels can also be automatically controlled.

Heat extraction and temperature control in the fluidized-bed combustor are accomplished by vaporizing demineralized water in 10 independent loops located in discrete vertical zones of the reactor. The water flows to these loops are controlled by valves whose positions change to maintain bed temperature in each of the zones.

Coal and makeup limestone to the combustor are fed continuously from a system designed for controlled solids feeding under pressure. Solids transfer between reactors and discharge of solids from the system (i.e., from the regenerator reactor) are accomplished using a pulsed gas transport technique controlled by pressure differentials across and between these fluidized beds.

INTRODUCTION

The fluidized-bed coal combustor provides a new boiler technique where coal is combusted in a bed of particles maintained in a state of fluidization by the air required for combustion. The use of limestone or other suitable sorbent as the bed material in such a system permits the capture and removal of sulfur dioxide simultaneously with the combustion process.

Fluidized-bed boilers (FBB) offer the potential of an efficient and compact boiler combustion technique also capable of providing pollution control. Some of the advantages and economic factors are:

1. Capability to combust lower quality fossil fuels in a fluidized bed.
2. Immersion of the boiler tubes directly in the fluidized bed achieves improved heat transfer rates compared to conventional boilers.
3. The higher volumetric heat release rates in a fluidized-bed combustor will permit reduced boiler unit sizes.
4. Since efficient combustion can be achieved at comparatively low bed temperatures (i.e., 1500-1700°F), boiler tube corrosion and fouling should be reduced.

Pressurized fluid-bed combustion offers even greater benefits in size reduction, efficiency, and load control.

Within the fluidized-bed boiler, limestone is calcined to lime which reacts with sulfur dioxide and oxygen in the flue gas to form calcium sulfate. When used on a once-through basis, relatively high limestone feed rates are required to the fluidized-bed boiler if sulfur dioxide removal in excess of 90 percent is to be maintained.

In order to reduce these high limestone feed rates, a system was proposed by Esso Research and Engineering whereby the calcium sulfate formed would be regenerated back to calcium oxide in a separate fluidized-bed reactor (i.e., regenerator) by reaction with a reducing gas at a temperature of approximately 2000°F. The regenerated lime would be returned to the fluidized-bed combustor, where it would again react with the sulfur dioxide.

In a study completed by Esso for the National Air Pollution Control Administration,[1] the following essential features of the proposed regenerative-limestone FBB system were demonstrated:

1. Removal of over 90 percent of the SO_2 formed by combusting coal in fluidized beds of lime.
2. Reductive regeneration of the sulfated lime to yield an off-gas containing 7 to 12 mole percent SO_2. This is a sufficiently high concentration to permit its conversion to H_2SO_4 or elemental sulfur with conventional technology.
3. Good activity maintenance of the lime cycled back and forth between combustion and regeneration. The make-up requirement for fresh limestone in a commercial plant was estimated to be about 15 percent of that required for once-through use of this material.

These experimental results were obtained at atmospheric pressure conditions. Since completing this study, engineering and cost analyses carried out by Westinghouse[2] for the Environmental Protection Agency (EPA) have indicated a much greater commercial potential for a pressurized FBB system when used in conjunction with a combined gas-stream turbine power generating plant. Based on this evaluation, the Office of Air Programs of EPA requested Esso Research and Engineering Company to design and construct a continuous fluidized-bed combustion-limestone regeneration pilot unit capable of operating at pressures up to 10 atm.

DESIGN BASIS

The parameters used for the design of the fluidized-bed miniplant are summarized in Table 1.

Table 1. DESIGN BASIS FOR FLUIDIZED-BED MINIPLANT

	Combustor	Regenerator
Unit dimensions		
Internal diameter, inches	12.5	5
Height, ft	28	19
Operating conditions (maximum)		
Temperature, °F	1750	2000
Pressure, atm	10	10
Superficial bed velocity, ft/sec	10	5
Heat released by combustion, Btu/hr	6,300,000	--
Cooling load	3,600,000	--
Material rates (maximum)		
Air, scfm	1250	90
Coal, lb/hr	480	--
Limestone, lb/hr	70	--
SO_2 generated, lb/hr	4	40

The limiting operating conditions for the combustor were set at 10 atm, 1750°F bed temperature, and a superficial bed velocity of 10 ft/sec. These maximum design conditions were based upon the engineering and economic analyses that had been carried out by the Westinghouse Research Laboratories.[2]

A 12.5-in. diameter combustor size was selected as a basis for the design because this would provide a system which could be constructed at reasonable cost and within reasonable time while still providing the essential data needed for future development of the pressurized fluidized-bed combustion technology. At the design conditions, a maximum coal feed rate of 480 lb/hr would be possible when operating the combustor with 15 percent excess air. With a heating value of approximately 13,000 Btu/lb, this coal rate would correspond to a heat release rate of 6.3×10^6 Btu/hr and require the removal of 3.6×10^6 Btu/hr of heat by the combustor cooling tubes to maintain a 1600°F temperature in the combustor.

The internal diameter and operating velocity of the regenerator were the next parameters to be specified. These are not independently adjustable parameters, but are related to each other and to the diameter and bed velocity of the combustor by a sulfur material balance over the system (Figure 1).

A critical factor in this balance is the sulfur dioxide concentration of the regenerator off-gas. Using an anticipated assumed value of 4 mole percent SO_2 concentration as the basis, a 5-in. (internal) diameter reactor operated at 5 ft/sec maximum superficial bed velocity was selected as the best compromise for the design of the fluidized-bed miniplant regenerator.

A material balance for the fluidized-bed miniplant operating at its maximum design coal feed rate (i.e., maximum conditions) is shown in Figure 2.

MINIPLANT DESCRIPTION

The overall system flow plan for the fluidized-bed miniplant is presented in Figure 3. Figure 4 shows the assembly drawing arrangement of the major miniplant components.

Main fluidizing air for the combustor and regenerator is supplied at operating pressures to 125 psig by a stationary compressor with a capacity of 1300 scfm. The air flow rates are measured by orifice flow meters and regulated by differential pressure transmitters and control valves. The superficial bed velocity in the combustor and regenerator can be controlled automatically and independently in this manner. In the combustor, the air passes through the distributing grid, up through the fluidized bed of solids, and out through two stages of cyclone for solids removal before it is cooled in a heat exchanger. The pressure in the combustor is maintained at a desired set point by a control valve in the exhaust line positioned by a pressure transmitter and controller.

Air for the regenerator can be electrically preheated for temperature control before passing into the reducing gas generator located at the bottom of the reactor. The reducing gas passes through a ceramic distributor plate which supports the fluidized bed. The exit gas from the regenerator is cooled by a heat exchanger and discharged through a control valve. The pressure in the regenerator is maintained about equal to the pressure in the combustor by a differential pressure transmitter and controller between the combustor and regenerator serving to position the control valve in the regenerator off-gas line.

Heat extraction and temperature control in the fluidized-bed combustor is accomplished by vaporizing demineralized water in 10 separate loops located in discrete vertical zones of the reactor. The water flow to these loops is controlled by valves whose positions automatically change to maintain bed temperature. The steam generated in these loops is condensed and returned to a reservoir.

Solids transfer between reactors and discharge of solids from the system (i.e., the regenerator reactor) are accomplished using a pulsed air transport technique controlled by pressure differentials across and between these fluidized beds. Coal and makeup limestone to the combustor are fed continuously from a system designed for controlled solids injection under pressure.

Combustion and Regeneration Reactors

The combustion and regeneration reactors constitute the heart of the FBCR Miniplant design. The combustor (Figures 5, 6) consists of a 24-in. steel shell refractory lined to an actual internal diameter of 12.5 inches. The overall height of 28 feet was chosen to provide a bed outage (i.e., dilute phase above the bed) at least equal to the expanded bed height that would be obtained at the maximum operating conditions. The reactor is designed in flanged sections, with a bottom plenum for the combustion air, and an upper section for discharging the flue gas to the cyclone.

The regenerator reactor (Figures 7, 8) consists of an 18-in. shell refractory lined to 5-in. ID. An overall reactor height of 19 feet provides for bed expansion and reactor outage.

Bed Support and Gas Distribution Grids

Figure 9 provides the details of the combustor grid design. This grid consists of 3/8-in. stainless steel plate containing 137 fluidizing nozzles on a 15/16-in. square pitch. Each of the 5/8-in. diameter fluidizing nozzles contains eight horizontal equally-spaced 5/64-in. holes. The combustor grid has been designed to provide a pressure drop of about 19 in. H_2O.

The regenerator grid is a high alumina cast ceramic disc that will be sandwiched between the flanges of the main regenerator and the bottom plenum. Disc orifice arrangement and sizing will give a pressure drop close to that of the combustor grid.

Cyclones and Discharge System

In the FBCR Miniplant design, flue gases and entrained solid particles from the combustor enter a two-stage cyclone separator system. The solid particles separated in the first stage cyclone are returned to the combustor near the grid via a dip leg extension pipe. Solids escaping the primary cyclone enter the more efficient second stage cyclone where they are separated and discarded by a lock hopper system. This technique permits the selective removal of fly ash and limestone fines from the system on a continuous basis.

Gas exiting from the regenerator enters a single stage cyclone, where the entrained particles are collected and discarded by a two-vessel discharge system. All cyclones are lined with refractory insulation and rated for operation at pressures to 10 atm.

Discharge gases from the cyclones are cooled in heat exchangers to reduce the exit gas temperatures. This minimizes the need for refractory lined pipe leading to the scrubber, and lowers the temperature rating required for the reactor back pressure control valves.

Combustor Heat Removal

At the maximum operating conditions for which the FBCR Miniplant has been designed, a combustor cooling load of approximately 3.6 x 10^6 Btu/hr is required to maintain a 1700°F bed temperature. Since the design calls for a

15-ft expanded bed height, 0.24 x 10^6 Btu/hr-ft of expanded bed must be removed. The design that has been developed for this purpose calls for control of bed temperature by water circulation through 10 individual serpentine tube loops located in discrete vertical zones of the expanded bed. Each loop occupies 18 inches of bed height and consists of 3/4-in. OD stainless steel tubes on a 2-1/4-in. horizontal pitch (Figure 10). The surface area of each tube loop is approximately 7.5 ft^2; it has been sized to handle the anticipated heat load (with two-phase flow in the tubes) assuming an overall heat transfer coefficient of 35 to 40 Btu/hr-ft^2-°F.

The coolant enters and exits the combustor through 5 special coolant distributor plates sandwiched between flanges at 3-ft vertical increments in the lower portion of the reactor. This arrangement obviates penetrating the refractory lined shell of the reactor and provides a means of combustor disassembly for inspection and maintenance.

The combustor cooling water is pumped from a feedwater storage tank through the fluidized-bed combustor tube loops, where a portion of it is vaporized. The liquid-vapor mixture then flows through a surface condenser where it is condensed and returned to the feedwater tank. Thus, the steam and saturated water generated in the combustor cooling tubes is condensed, cooled, and recirculated to the combustor to maintain a clean, closed cooling water system. The fresh make-up water required is demineralized before entering the feedwater storage tank. This recirculating arrangement is intended to minimize cooling tube fouling, thereby maintaining effective heat transfer and extended tube life.

The design includes a technique for determining the heat transferred to the two-phase cooling water/steam system. Terminal temperature measurements at the cooling coil, flow rate measurements of the feedwater to the cooling tubes, and the use of throttling calorimeters at the coil exits provide data to permit the calculation of the heat transferred to the combustor cooling coils. These data can be used to determine the overall heat transfer coefficient for the coil in the fluidized bed.

Coal and Limestone Injection

The design of the coal and limestone injection system for this fluidized bed miniplant has been provided by Petrocarb, Inc. Petrocarb states this system to be capable of continually feeding the required mixture of coal and limestone to the combustor at a rate of 550 lb/hr against a combustor pressure of 10 atmospheres. Petrocarb claims that the coal must have negligible surface moisture for reliable injection operation.

Esso Research and Engineering will purchase coal that is suitably crushed and dried, and load this in a hopper/conveyor where it is delivered to a 15-ton coal storage bin. Limestone is handled similarly and stored in an adjacent 2-ton capacity storage bin.

Volumetric feeders deliver coal and limestone in the ratio of approximately 5 to 10 parts of coal to 1 part limestone from their respective storage bins to a blender and then to a feed injector vessel. This mixture of coal and limestone is then transferred pneumatically to the primary injector vessel upon a demand weight signal from the primary injector.

After the charge is transferred from the feed injector to the primary injector, the feed injector is isolated, vented, and refilled in preparation of a new weight demand signal from the primary injector. The weight cell on which the primary injector is mounted is also used to monitor and control the materials feed rate to the combustor.

Aerated solids in the primary injector gravity flow through an orifice into a mixing section where a controlled air stream transports them into the combustor. The solids feed rate is regulated by the transport air flow rate and the pressure difference between the primary injector vessel and the combustor. This feed rate is automatically controlled by the load cell on the primary injector and a rate controller which senses and adjusts the rate of mass decrease in the injector vessel.

Solids Transfer

Stone is continuously transferred from the combustor to the regenerator and from the regenerator to the combustor by inducing the solids to surge into an overflow reservoir immersed in the upper expanded bed of the reactor. The solids then flow (by gravity) down the transfer lines into receiving pots located near the grids of the two reactors. From these lower receiving pots (Figure 11), solids are entrained and transported into the reactor by timed and metered pressurized nitrogen pulses. The pulse interval, frequency, and nitrogen flow rate regulate the rate at which these solids are transferred. Excess solids for discard are also removed from the regenerator by this technique.

Bed Level Control

Since the pressure drop across a fluidized bed is directly proportional to the weight of solids in that bed, the design incorporates a differential pressure transmitter circuit to measure and control the amount of material in the combustor and regenerator reactors, their bed levels at the particular fluidizing conditions. Stone transfer to control bed levels is achieved by adjustment of the on-cycle operation of the pulse feeder mixing chambers. The regenerator mixing chamber is intended to be pulsed continuously, but the chamber returning solids to the combustor will operate only when the stone inventory in the regenerator exceeds its set value. This regenerator value increase is sensed by the differential pressure cell as an increase in pressure drop across the bed; the pulse air flow solenoid valve will open to transfer solids from regenerator to combustor.

The stone discard flow from the regenerator, and therefore the total reactor system solids inventory will be controlled similarly. Solids will be discarded from the regenerator to an air cooled receiving reservoir when the pressure drop across the combustor bed and the regenerator bed both indicate high levels. By this technique, the bed levels in both the combustor and regenerator can be controlled by automatically regulating the solids transfer and discard rates.

The regenerator receiving reservoir will also serve to receive the solids from the regenerator cyclone. From this reservoir, the cooled solids will be transferred periodically to a second vessel capable of being depressurized for solids removal.

Reducing Gas Generator

A reducing gas generator capable of producing 10,000 scfh of gas at 150 psig supplies reducing gas to the regenerator. The unit is a 24-in. OD carbon steel cylinder internally insulated to create an 8-in. ID combustion chamber. The insulation is cast to form a 5-in. diameter discharge nozzle and an off-set shoulder for mating with the regulator to provide a means for installing a distribution plate and radiation shield.

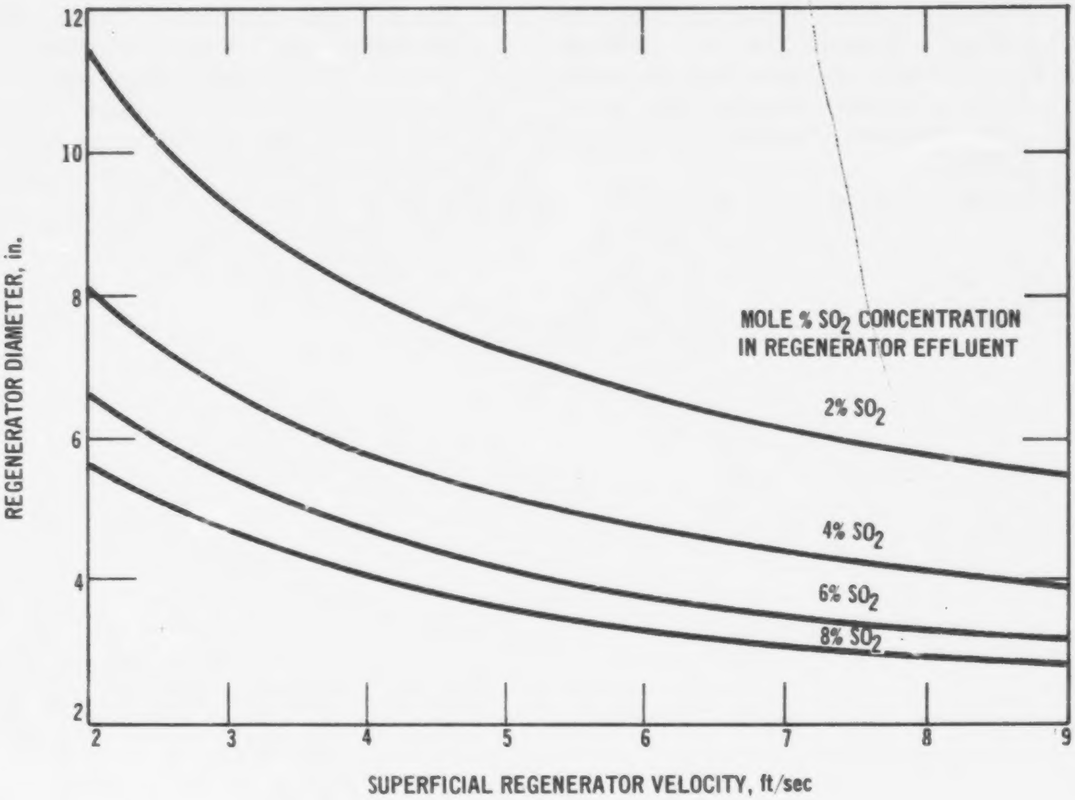
The gas/air burner gun is AISA type 309 SS and enters via a combustion-air-cooled nozzle while the pilot enters via a 2-in. flanged nozzle on the chamber side. Two observation windows (2-in. flanged with quartz windows) are provided to allow flame viewing: one for visual and one for infrared scanner use. Pilot and main flame monitoring is by electric scanner of weather-proof construction with flame detector amplifier and relay used to actuate a gas solenoid valve. A high tension electric spark for pilot ignition is provided by a 100V/10,000V transformer.

REFERENCES

1. Hammons, G. and A. Skopp. A Regenerative Limestone Process for Fluidized Bed Coal Combustion and Desulfurization, Final Report. Esso

Research Centre, Abingdon, Berkshire, England. Prepared for Air Pollution Control Office, Environmental Protection Agency, Research Triangle Park, N. C. under Contract Number CPA 70-19. February 1971.

2. Archer, D. H., et al. Evaluation of the Fluidized Bed Combustion Process. 15th Monthly Progress Report. Westinghouse Research Laboratories, Pittsburgh, Pa. Prepared for the Air Pollution Control Office, Environmental Protection Agency, Research Triangle Park, N.C. under Contract Number CPA 70-9. March 1971.



Above curves have been developed from the equation:

$$\frac{T_R}{T_C}\cdot\frac{C_C^{SO_2}}{C_R^{SO_2}}\cdot V_C D_C^{2} = V_R D_R^{2}$$

which is based on a SO_2 material balance between the combustor and regenerator. In this equation,

T = Absolute temperature

C^{SO_2} = SO_2 concentration (based on S content of coal for the combustor)

V = Superficial velocity

D = Reactor diameter

and the subscripts refer to the regenerator conditions (R) and the combustor conditions (C).

Figure 1. Relationship between regenerator diameter and operating velocity.

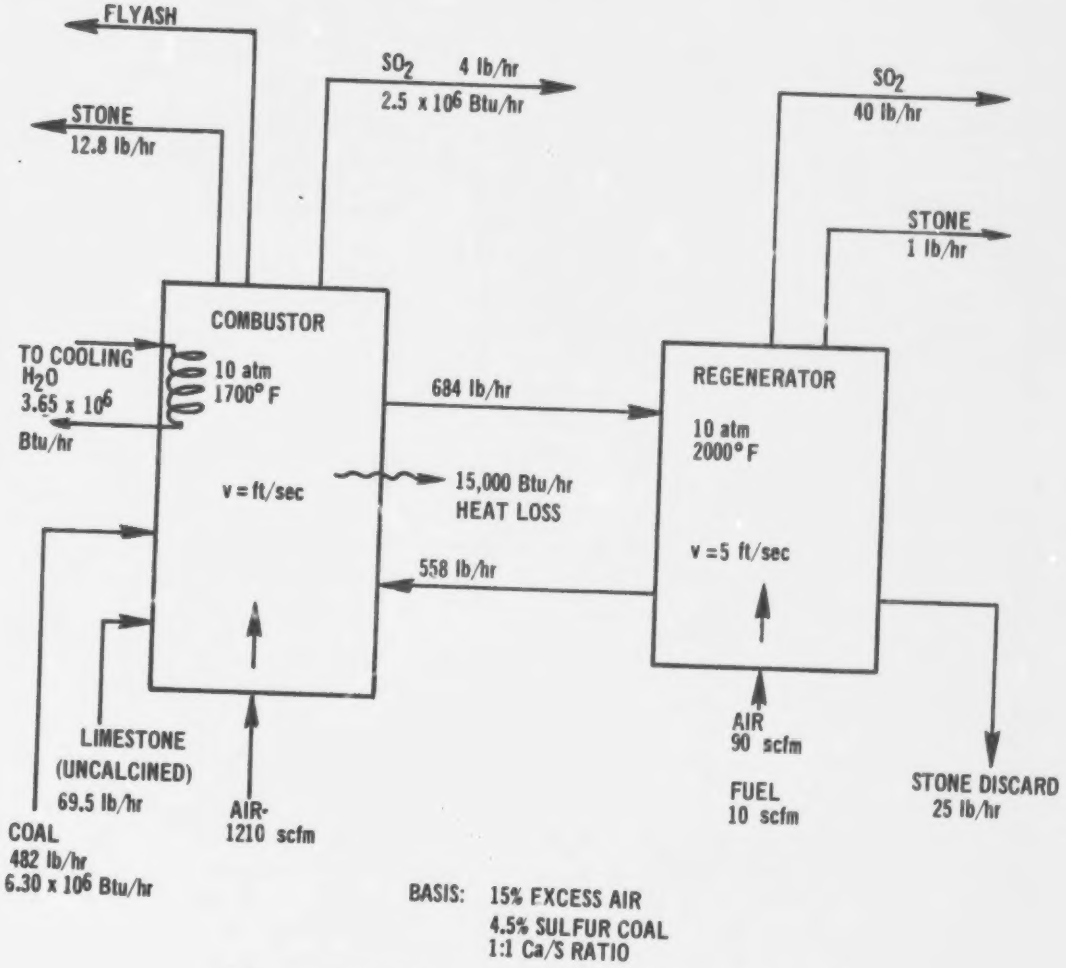


Figure 2. Material balance for the FBCR miniplant.

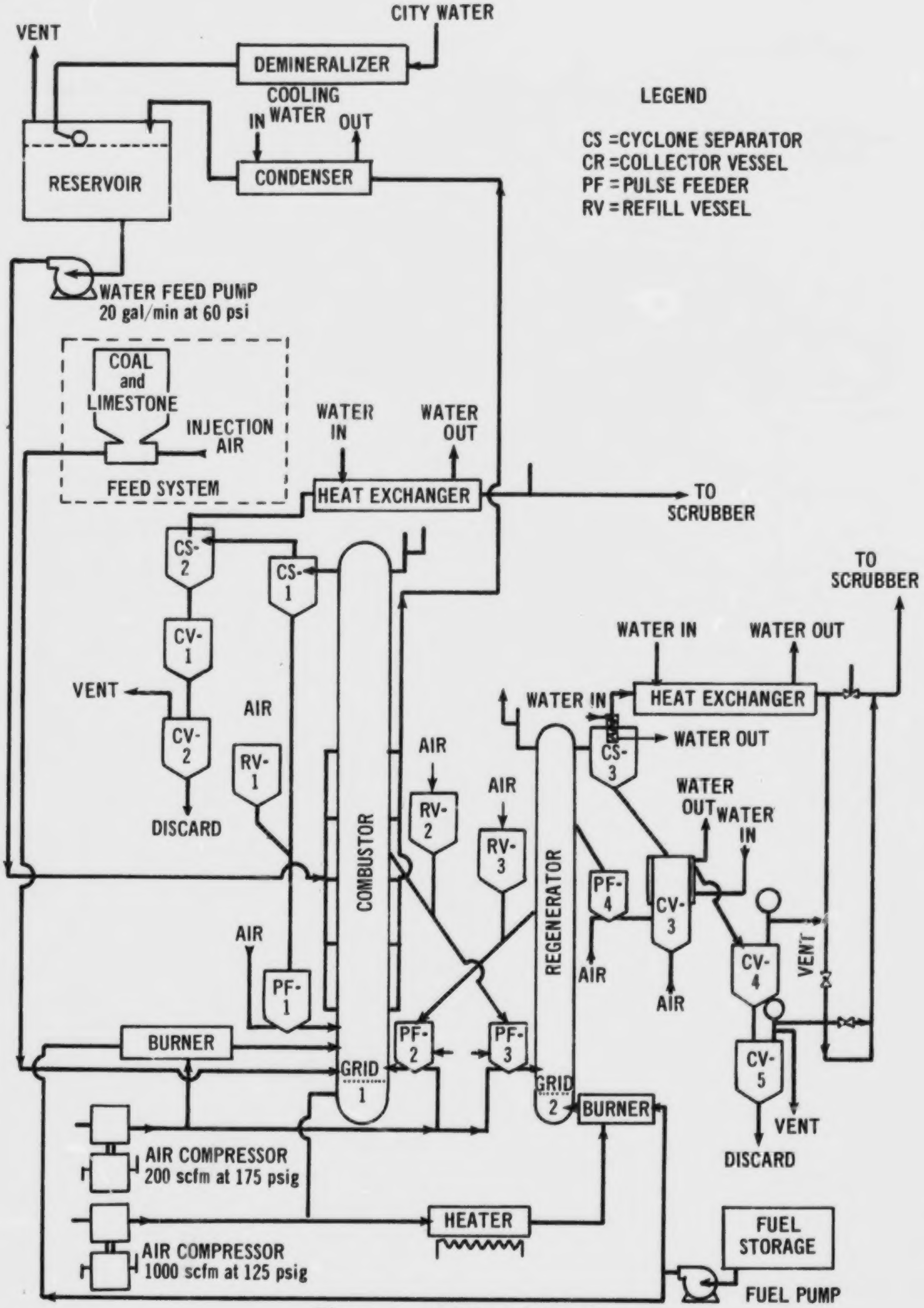


Figure 3. FBCR miniplant flow plan.

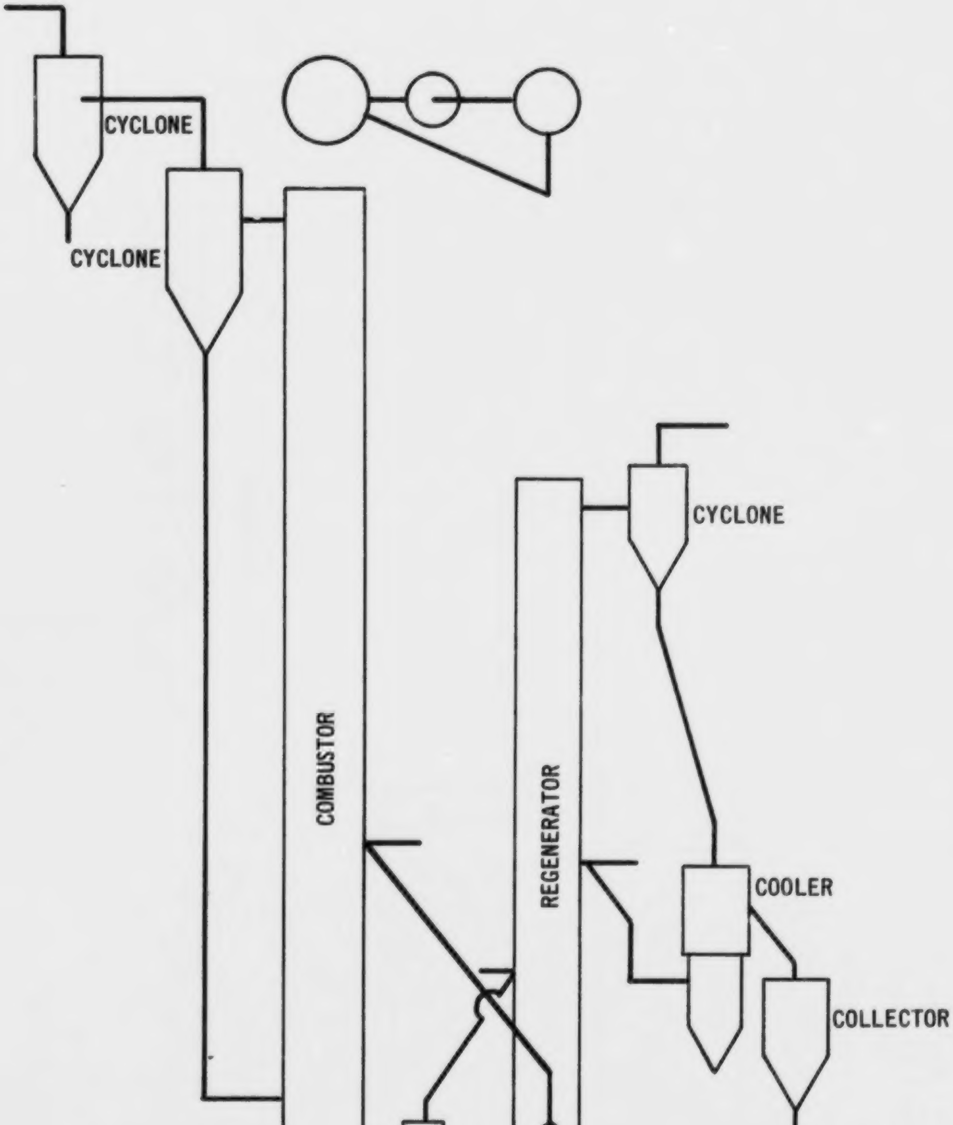
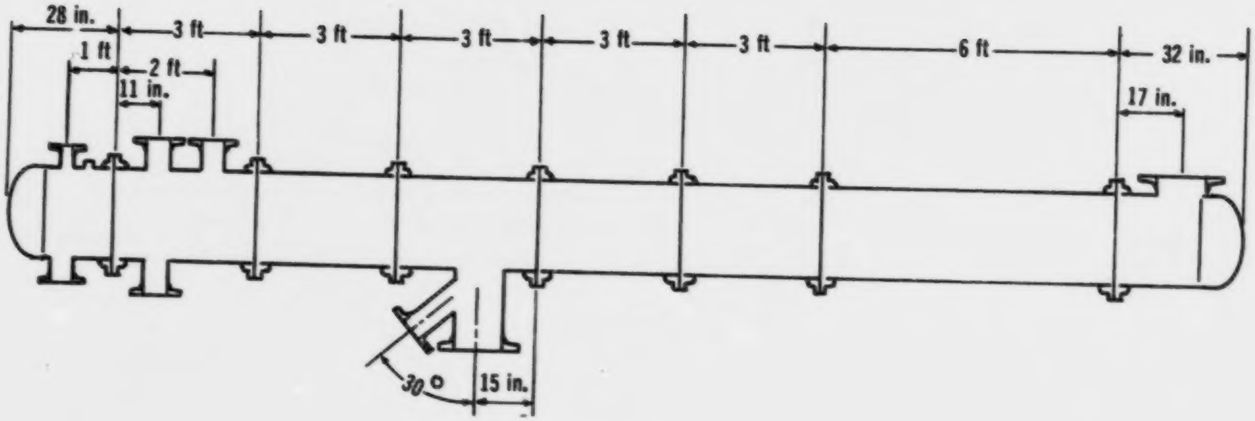


Figure 4. FBCR miniplant assembly.



TO BE TESTED AND A.S.M.E. CODED
FOR 150 psia WORKING PRESSURE
AT 400°F.

MATERIALS

SHELL - 24 BY 0.375 in. WALL STEEL PIPE
SHELL FLANGES - 24 BY 0.150 in. STEEL R.F. SLIP-ON
SHELL ENDS - 24 in. STANDARD WALL WELD CAPS

Figure 5. Combustor shell.

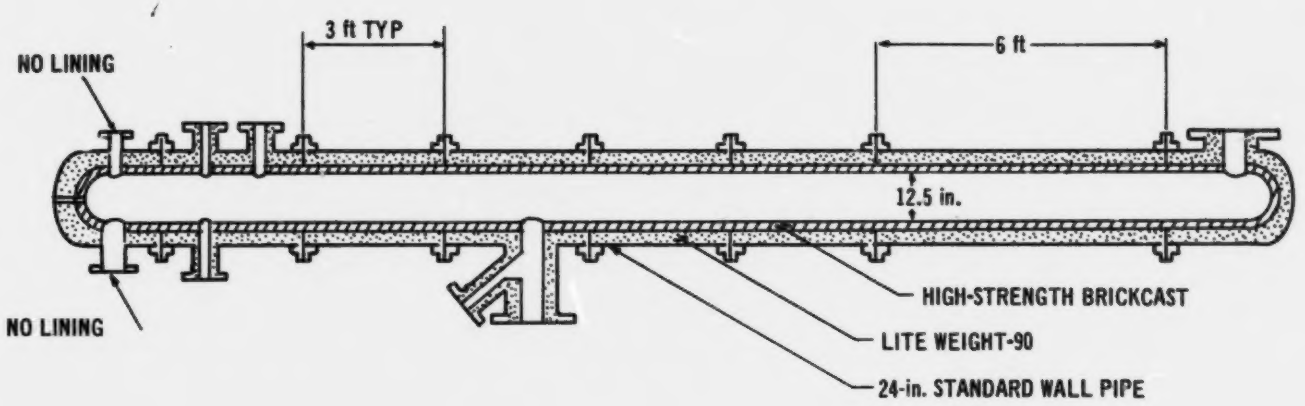
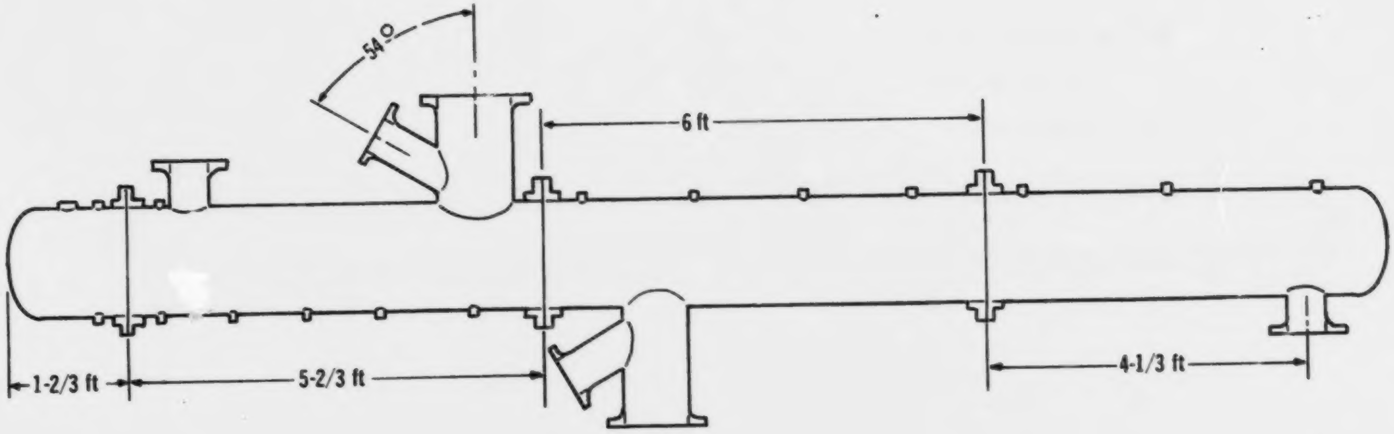


Figure 6. Refractory lined combustor.



TO BE TESTED AND A.S.M.E. CODED
FOR 150-psia WORKING PRESSURE
AT 400 °F

MATERIAL

SHELL - 18 BY 0.375 in
SHELL FLANGES - 18 BY 0.150 in.
SHELL ENDS - 18 in. STD WALL WELD CAPS

Figure 7. Regenerator shell.

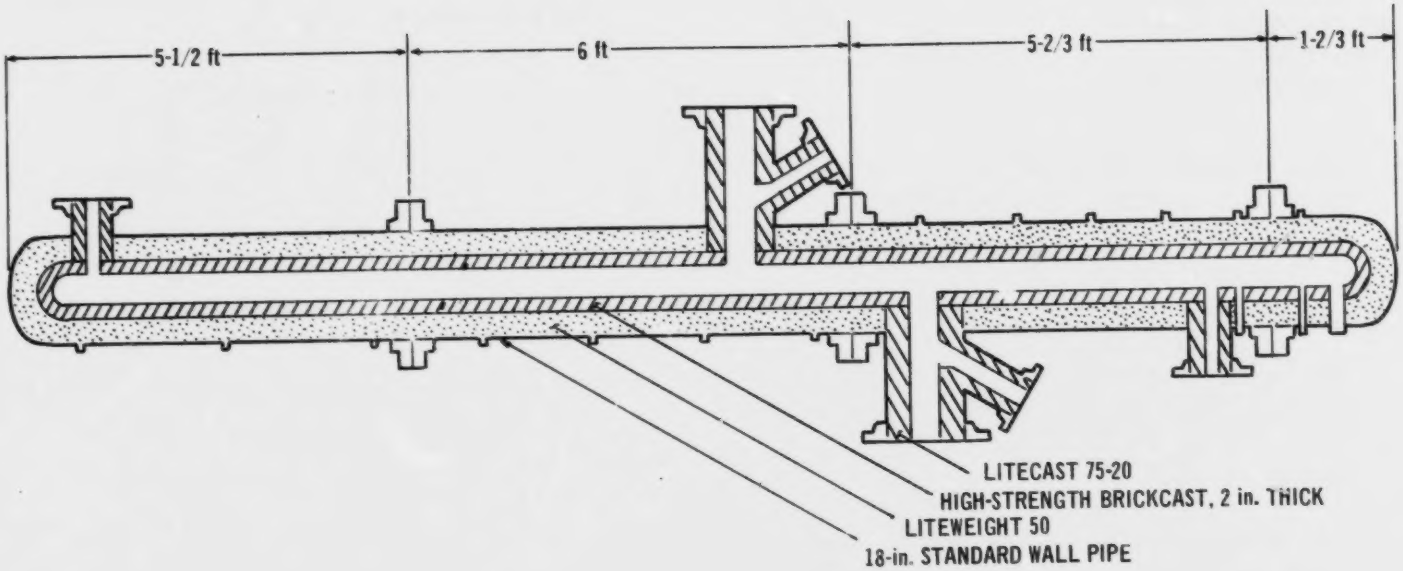

5-1/2 ft
6 ft
5-2/3 ft
1-2/3 ft
LITECAST 75-20
HIGH-STRENGTH BRICKCAST, 2 in. THICK
LITEWEIGHT 50
18-in. STANDARD WALL PIPE

Figure 8. Refractory lined regenerator.

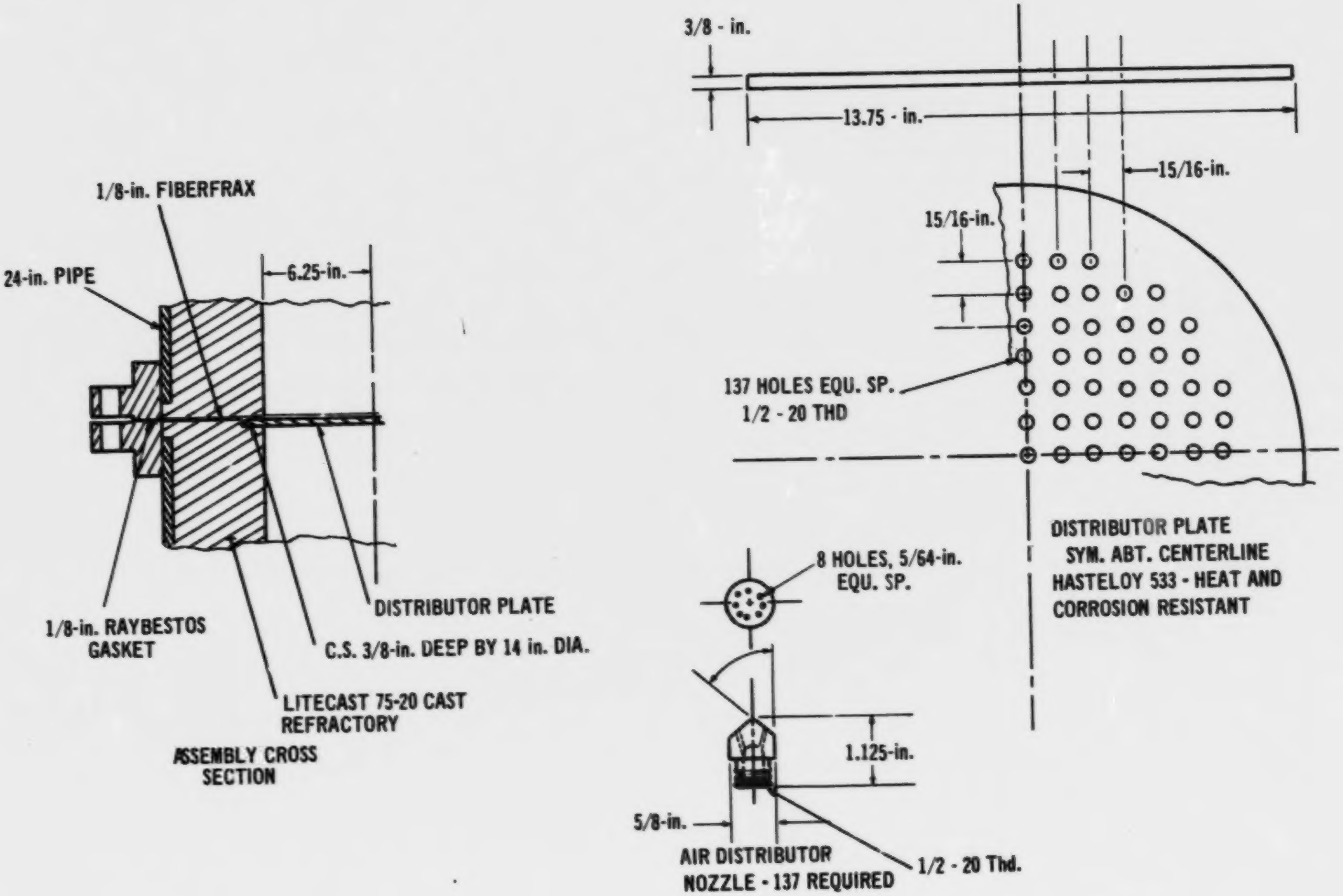


Figure 9. Combustor air distributor plate.

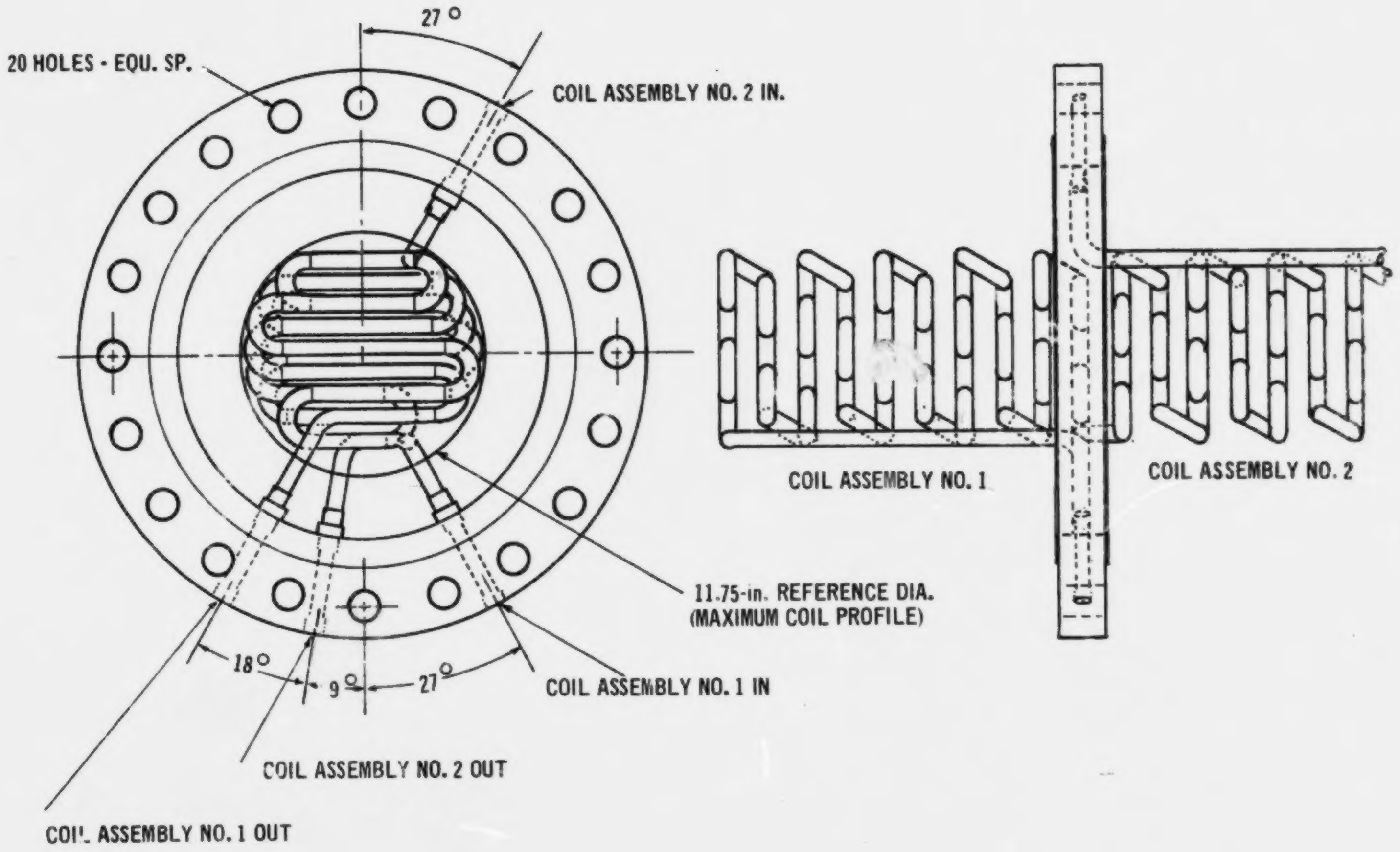


Figure 10. Combustor cooling coils.

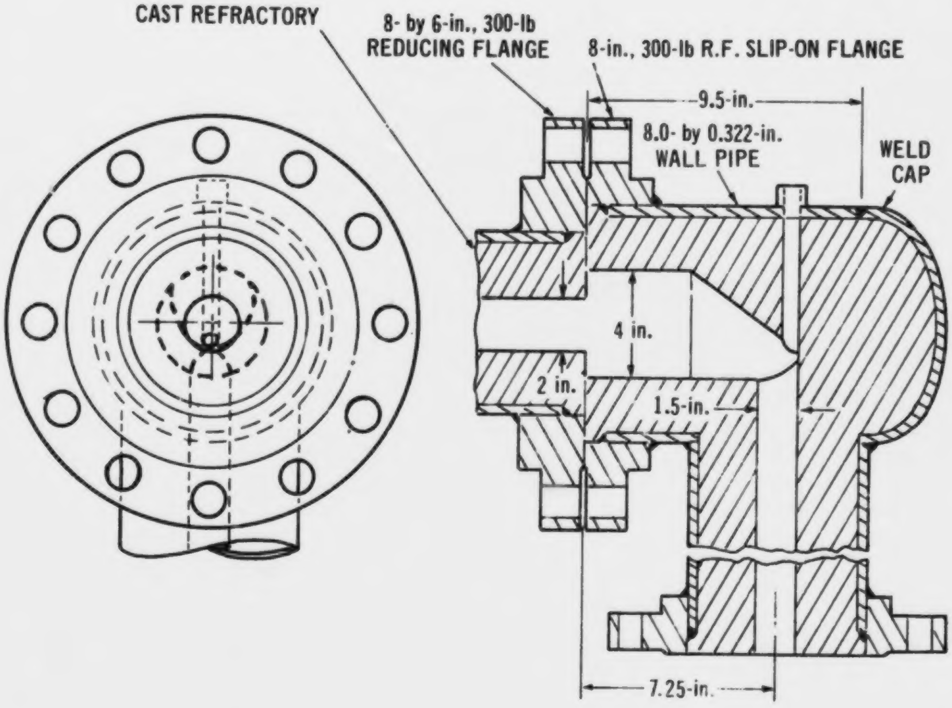


Figure 11. Pulsed solids transfer pot.

3. A PRESSURIZED FLUIDIZED-BED BOILER DEVELOPMENT PLANT

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and
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ABSTRACT

Preliminary designs have been prepared for a 30-MW pressurized fluidized-bed combustion boiler development plant. Such a plant is needed to further the development of a power generation concept which promises reduced costs for electrical energy and reduced emissions of SO_2, NO_x, and particulate pollutants from the use of coal and oil. The designs—together with an experimental program, schedule, and budget—will be useful in:

1. Focussing on technical problems involved in developing commercial fluidized bed combustion power plants.
2. Planning the development.
3. Forming a development team.

Information received from laboratory and pilot plant during 1973 and 1974 can be used to improve the development plant equipment design and experimental program. The formation of a team to implement the development plant program should be accomplished during 1973.

Detailed design, construction, and operation of this plant are recommended for the 3-year period 1974-1976 in order that a demonstration pressurized fluidized-bed combustion boiler power plant can be built and operated before the end of the decade.

INTRODUCTION

A recent technical and economic analysis[1] has shown that power plants using pressurized fluidized-bed boilers can be built which are:

> Economic in capital requirements—Estimated costs for such plants are 20-30 percent less than for conventional steam power plants.

> Efficient in operation—Pressurized fluidized-bed boiler plants using conventional steam technology are equal in overall efficiency to the best of conventional plants—about 38 percent: the use of increased steam temperatures and pressures and of increased combustion gas temperatures will increase overall plant efficiency to 46 percent or more.

Effective in pollution abatement—Pressurized fluidized-bed boiler power plants meet emission requirements established for SO_2, NO_x, and particulates.

A preliminary design has been prepared for a 30-MW pressurized fluidized-bed boiler development plant. The boiler comprises a single fluidized bed of 5 x 7 ft rectangular cross-section. Bed depths of 6 to 35 feet are needed to accommodate tubes for steam generation. The boiler represents a single bed in a module of a 300 to 400-MW power plant boiler.

Such a boiler is required to obtain data for the design, construction, and operation of a demonstration power plant based on pressurized fluidized-bed combustion. The preliminary plans provide a basis for detailed designs and for overall planning through the preparation of site requirements, program schedules, and cost estimates. The plans will be useful also in gathering technical ideas for the development plant and in focusing attention on the technical problems which it must solve.

After costing of the design has been completed, the formation of a U.S. government, industry, and utility team will be required to undertake the financing, construction, and operation of the development plant. This plant should be operating in 1975-1976 in order to provide the necessary data for a demonstration pressurized fluidized-bed combustion boiler power plant to be constructed by 1980.

BACKGROUND INFORMATION

Concept

A 635-MW pressurized fluidized-bed combustion boiler power plant has been designed.[1] The power cycle schematic is shown in Figure 1. Operating at elevated pressure, a fluidized-bed combustor requires a compressor to pressurize the air and to overcome the pressure loss over the fluidized-bed combustor. At an operating pressure of 10 to 15 atm, excess air of 10 to 15 percent and fluidizing velocity of 6 to 12 ft/sec, a depth of 8 to 15 feet is required to accommodate the heat transfer surface in the bed; the pressure loss over the bed is thus 4 to 8 times as large as that over an atmospheric-pressure bed. The pumping energy, however, is actually less because of the greater density of the gas at high pressure. Energy can be recovered from the high-temperature, high-pressure gases by passing them directly into a gas turbine expander, reducing their pressure to atmospheric as shown in Figure 1. This expansion lowers the temperature of the gases by 600 to 800°F, thereby reducing the amount of surface required to recover heat from the combustion gases leaving the fluidized bed. The pressurized system can be operated at higher excess air rates. Such operation increases the fraction of gas turbine power, reduces combustible losses from the boiler, and increases the waste heat recovery after the gas turbine. It also results in improved plant efficiencies.

Several boiler systems have been built, tested, or proposed which incorporate fluidized-bed combustion. These systems, as well as alternative concepts, have been evaluated. The early concepts did not incorporate heat transfer surface or sulfur removal in the bed and are generally designed to burn low grade fuels. The heat released was extracted from the combustion gases during their passage through a conventional boiler. Recent concepts and studies incorporate heat transfer surface in the bed to achieve a more compact and efficient design and/or remove sulfur during the combustion process by using a limestone bed.

The boiler design considered most promising consists of four modules; the modularized design provides for a maximum of shop fabrication and standardization and assists in meeting the turn down requirements for the plant. Each module includes four primary fluidized-bed combustors, each containing a separate boiler function—one bed for the pre-evaporator, two beds for the superheater, and one bed for the reheater. Evaporation takes place in the water walls. All of the boiler heat transfer surface is immersed in the beds, except for the water walls. There is no convection heat transfer surface since the maximum allowable bed temperature is less than the state-of-the-art gas turbine temperature. The fluidized-bed combustors are stacked vertically because of advantages in gas circuitry, steam circuitry, and pressure vessel design to achieve deep beds. Each module contains a separate fluidized-bed or carbon burn-up cell (CBC) to complete the combustion of any carbon elutriated from the primary beds. A separate bed may not be required since carbon losses may be low enough in the proposed pressurized boiler design with deep beds. A CBC is not envisaged if the system is operated with high excess air. This has the attraction of increasing plant performance by increasing the fraction of gas turbine power A simplified drawing of a module is shown in Figure 2. The 318-MW plant module is rail-shippable and can be shopfabricated. The 635-MW plant module is designed to be shipped in sections, each shop-fabricated. The primary beds for a 318-MW plant are approximately 5 x 7 ft. The bed depths are approximately 12 feet—sufficient for the required heat transfer surface. The CBC is approximately 2 x 7 ft; it contains no submerged surface in the bed. The submerged tube bundles are formed by vertical tube platens or planes; each platen is a continuous boiler tube in a serpentine arrangement. A platen is schematically represented in Figure 2. The heat transfer surface can be viewed as horizontal tubes. The pre-evaporator and reheater contain 1-1/2-in. diameter tubes; the evaporator water walls and reheater bed contain 2-in. diameter tubes. Details of the boiler design and the plant layout are presented elsewhere.[1]

The effectiveness and economics of a pressurized fluidized-bed boiler power plant have been evaluated.[1] Demonstrated SO_2 and NO_x reductions are adequate to meet emission standards. Energy costs are projected to be ~ 10 percent less than conventional power plants using current power generation technology. There is thus a large economic margin for solving technological problems; there is also potential for increasing performance and reducing costs by increasing both gas turbine and steam turbine performance.

The primary potential advantages of a fluidized-bed boiler power plant are:

Reduced volume and modular construction — Because combustion rates are more intense in fluidized beds than in the fire box of a pulverized fuel furnace, and because heat transfer surface can be placed in the bed, fluidized-bed boilers are more compact than conventional coalfired boilers. Pressurized boiler modules can be fabricated in shops and installed at a power plant site. Considerable economics are possible in the fabrication and erection of pressurized fluidized-bed combustion boilers.

Reduced heat transfer surface requirements — Because heat transfer coefficients are in order of magnitude greater in fluidized-beds than in the fire box of a conventional boiler, less heat transfer surface is required in the boiler. In a pressurized boiler, less heat is extracted from the combustion gases after they leave the fluidized-bed because they are cooled by expansion in the gas turbine. Further large reductions in heat transfer surface are thus possible. The heat transfer surface in the pressurized boiler is ~ 80 percent less than the surface in a conventional pulverized fuel boiler.

Reduced steam tube and turbine blade corrosion, erosion, and fouling—Because the fluidized-bed boiler operates at a maximum combustion temperature far below that in a conventional boiler, volatilization of alkali metal compounds and fusion of ash is reduced or eliminated. Sulfur and vanadium compounds are removed from the combustion gases by the sorbent. The corrosion/erosion and fouling of steam tubes and turbine blades is thus minimized. Under these conditions, higher steam temperatures and pressures may become economically feasible, and greater efficiency in power generation may be achieved. Somewhat greater efficiencies can also be achieved in pressurized fluidized-bed combustion by using gas turbines with high inlet temperatures.

Reduced fuel costs and increased flexibility—Because fluidized beds can readily burn crushed coal (fine grinding is not required), coal with a high ash content, and a variety of miscellaneous combustibles (from sewage sludge, municipal solid wastes, paper mill liquid wastes, and oily wastes to residual oil and natural gas), boilers using such beds can utilize cheap fuels and a wide variety of fuels to generate power and steam.

Reduced emissions of SO_2 and NO_x—Because a limestone/dolomite sorbent can be utilized in a fluidized-bed combustor, SO_2 reductions of 90 to 95 percent can be economically achieved. The low combustion temperature at which the bed operates minimizes the formation of NO_x by fixation of atmospheric nitrogen. Production of NO_x by oxidation of nitrogen in the fuel is minimized by operating the bed at high pressure. Compliance with NO_x emission regulations at atmospheric pressure will require design and/or operating modifications to the boiler.

Several characteristics of a pressurized fluidized-bed boiler must be demonstrated:

operation of deep beds with internals, adequate particulate removal for reliable gas turbine operation and sorbent regeneration or high stone utilization to permit once-through operation.

Pressurized fluid-bed boiler apparatus and support facilities have been operated to supply information on:

1. Combustion and combustion efficiency: carbon carry-over; burning above bed–carbon solids and gases.
2. Pollution abatement: SO_2 removal, regeneration, and recovery; NO_x reduction; particulate–formation, ash, sorbent attrition, and removal.
3. Heat generation and transfer: combustion in deep beds; temperature distribution; and heat transfer coefficient distribution.
4. Reactant feed and distribution: fuel, sorbent, and air.
5. Materials: steam tube and turbine blade.

This information can be obtained on different types of experimental apparatus and on different scales. An evaluation of these alternative apparatus are presented in Table 1. A summary of available and planned pressurized fluidized-bed boiler apparatus is summarized in Table 2.

PRESSURIZED FLUIDIZED-BED COMBUSTION BOILER DEVELOPMENT PLANT

The pressurized fluid-bed combustion boiler program at BCURA demonstrated the feasibility of pressurized fluid-bed combustion in an 8-ft^2 combustor. Specifically, it demonstrated:

SO_2 emissions of $\leqslant 0.7$ lb/10^6 Btu.
NO_x emissions of 0.07-0.2 lb $NO_2/10^6$ Btu.
Coal feeding at 3-1/2 and 5 atm.
Continuous operation—runs up to 350 hours.
No erosion or corrosion of gas turbine blade.
Test passages after 200-hour tests.
Particulate removal at ~ 1500°F and 5 atm by cyclones proved adequate for turbine blade tests.
Adequate boiler tube materials for commercial applications.
Bed operation with horizontal tube bundle.

Commercial boiler design operating conditions require that the bed be operated at higher gas velocity, higher bed pressure, higher temperature, and with a deeper bed than the BCURA unit. A development plant is required to investigate the design, construction, and performance of the proposed boiler plant equipment system design at the proposed operating conditions so that commercial feasibility might be assessed. Several features require a large plant to evaluate the design and performance.

Operation of deep beds (10 to 20 feet) with horizontal tube bundles and headers with aspect ratios $< \sim 2.5$.
Heat generation and temperature distributions.
Coal feeding requirements and sorbent distribution in a large bed (~ 35 ft^2).
Particulate control equipment performance.
Heat transfer surface configuration and materials requirements.
Gas turbine blade materials and component life.
Disengaging height design criteria.
Operational techniques—startup, shutdown, load follow, stability.

Long-term operability at design and operating conditions.
Fabrication, maintenance, and repair of boiler and auxiliary components.

The plant is also required to confirm on larger scale results obtained in laboratory and bench apparatus and in pilot plants.

Emissions—SO_2, NO, CO, C, alkali metals.
Combustion efficiencies.
Sorbent utilization.
Sorbent attrition.
Air distribution.
Boiler tube materials.
Particle carry-over and the sources.

A small scale unit will not provide solutions to the following problems:

1. Erosion that may occur due to tube configurations found only in a full scale unit.
2. Feed point location distribution requirements for coal and dolomite into a full size fluidized bed (number of feed points and their location).
3. Thermal inertia effects in turndown and startup that can be determined only from a full scale module.
4. Physical arrangement for maintenance access (tube-repair, coil replacement, instrument replacement, etc.) that can only be designed into a full scale module.
5. Determination of shop fabrication methods and costs, and the associated shipping protection requirements that only a full scale module would require.
6. Mechanical design methods suitable for tube bundle support, grid plate support, differential expansion of water ,wall penetrations, etc., that can only be proven adequate in a full scale module.
7. Investigation of tube vibration over the long span tube lengths only utilizable in a full scale module.
8. Determination of field erection methods and costs, not obtainable from a small scale unit.
9. Determination of various unusual operating effects on the full scale module mechanical design. Conditions created by an emergency shut-down of the turbo-expander, or a low flow surge of the centrifugal air supply compressor, would be used to test the design adequacy of the full scale module.
10. Investigation of optimum space utilization and heat transfer surface configuration within the fluidized bed can best be proved out by testing alternate tube bundle designs in the full scale module.

Other objectives which are being considered for the facility include:

Need to test sulfur recovery system.
Feasibility of studying advanced concepts—higher steam temperature and pressure, higher gas turbine temperatures, circulating beds, deeper beds (30) ft), higher pressures.
Feasibility of expanding to multiple bed operation.

The development plant is planned so that sufficient information can be obtained to design, build, and operate a demonstration plant of 150 to 300 MW.

Design Basis

The development plant design is based on the objectives outlined for it and for the commercial plant design. For the plant to provide sufficient information to design, build, and operate a demonstration power plant, the unit must be large enough to:

1. Test multiple point coal feeding.
2. Avoid untypical height/diameter ratios with bed operation.
3. Test proposed heat transfer surface configurations.
4. Test effect of bed emissions on gas turbine performance.

These design characteristics can be met by constructing a fluidized-bed unit with a capacity equivalent to one bed in a 75- to 100-MW_e module.[1] The range of operating conditions and design criteria established for the plant are summarized in Table 3.

Table 1. PILOT PLANT TYPES AND SCALES

A. Laboratory apparatus (TGA, DTA, etc. small fluidized beds)

Batch operation
Discrete operations
Sample sizes: 0.01 g -100 g
  Combustion—devolatilization (?), char oxidation (?)
  Abatement—SO_2 removal, sorbent regeneration
    NO formation (?) and extinction
    ash formation (?)

kinetics; extent of reaction; effects of T, p, x; understanding of basic phenomena; nature of solids

Time and cost of operation
  $5,000-$25,000 for equipment
  1/2 day/experiment, 1 technician, 1 engineer: $500/experiment

B. Bench apparatus (3-6-in. diameter fluidized beds)

Semi-continuous (fuel flow, limestone batch)
Discrete operations
Fuel flow: 10-100 lb/hr
  Combustion—C carryover
  Abatement— SO_2 removal, regeneration
    NO reduction, ash
  Materials—steam tube

interaction of phenomena; effects of combustion on abatement

Time and cost of operation
  $25,000-$100,000 for equipment
  2-3 days/experiment, 2-3 technicians, 1-1/2 -2 engineers: $3000/experiment

C. Pilot apparatus (12-36-in. diameter fluidized beds)

Continuous (fuel and limestone flow)
Integrated or discrete operation
Fuel flow: 100-1000 lb/hr
  Combustion—carryover and burning above bed
  Abatement— integrated operation (?), ash
    formation, limestone attrition
  Heat transfer— axial distribution generation
    and temperature, h value
  Materials—tube and blade

integration of heat transfer and regeneration, deep bed operating with submerged tubes

Time and cost of operation
  $500,000-$1,500,000 for equipment
  5-20 day/experiment $20,000-$100,000/experiment

D. Development (5-7-ft diameter fluidized beds)

Continuous
Integrated or discrete
Fuel flow: 10,000-30,000 lb/hr
  Combustion—carryover and burning above bed
  Abatement—attrition
  Heat transfer—full scale
  Reactant distribution—coal, limestone, air
  Materials—tube and blade under more realistic
    conditions

scale-up, full scale bed and tube arrangement, integration of feed and distribution phenomena, steam generator and materials examined under more realistic conditions. Equipment fabricating and operating problems.

Time and cost of operation
  $8,000,000-$15,000,000
  5-100 days/experiment

E. Demonstration (multi- 5-7-ft diameter fluidized beds)

Continuous
Integrated
Fuel flow: (80,000-120,000 lb/hr)

  All phases of a complete plant

overall system performance and costs demonstration

Time and cost of operation
  $30,000,000-$60,000,000 for equipment
  30-300 days/experiment

Table 2. PRESSURIZED FLUIDIZED-BED BOILER APPARATUS

Type	Location	Capacity		Operating limitations			Purpose	Status
		Diameter	lb coal/hr	Temperature, °F	Pressure, psi	Gas velocity, ft/sec		
Special purpose								
TGA	Westinghouse	-	-	2,400	450	-	Kinetic studies on sulfur removal/ regeneration	Operating
TGA	CCNY	-	-					
Fluid bed	Argonne	2 in.	-	>2,000	150			Operating
Fixed bed	Esso	1 in.	-	>2,000	150	5		Operated
Cold models	Westinghouse	1.5- x 9-in.	-	ambient	ambient	1-40	Alternative concepts	Operating
		4 in.	-	ambient	150	up to 6	Fluidization and solids handling	Operating
	Esso	4 in. ?	-	ambient	40	up to 10	Fluidization and solids handling	
Pilot plants								
Semi-batch	Argonne	6 in.	60	1,900	135		Sulfur removal/	Operating
	Argonne	3 in.	-	2,100	135		Combustion regeneration	Operating
Semi-batch	Esso	3 in.	-	2,200	150	5	Regeneration	Operating
	Esso						Combustion/sulfur removal	Nov. 1972?
Continuous	Esso	5 in.	-	2,000	150	5	Regeneration	Jan. 1974
		12.5 in.	480	1,700	150	10	Combustion/sulfur removal	Jan. 1974
Semi-batch	BCURA	2- x 4-ft	~400	1,500	90	2	Combustion/sulfur removal	Operated 1969-1971
Development plant								
	?	5- x 7-ft	nominal 22,000	2,000	225	15	Data for demonstration plant	Preliminary design, Dec.

Table 3. DESIGN BASIS FOR PRESSURIZED FLUIDIZED-BED BOILER DEVELOPMENT PLANT

Coal	Pittsburgh No. 8 (4.3% S)
Dolomite	
Pressure	1-20 atm
Temperature	1500-2000°F
Gas velocity	6-15 ft/sec
Bed area	~ 35 ft^2
Bed depth	4-30 ft
Ca/S	1-6 for once-through 2-10 for regeneration
Heat transfer coefficient	50 Btu/hr ft^2 °F
Particulate carry over	
Particle size	
Coal	1[illegible] x 0
Dolomite	1/4 in. x 0 or 1/4 in. x 28 mesh
Site	Existing power plant
Air supply	Separate air compressor, motor drive
Excess air	> 100% capability
Air preheat	to 600-850°F
Feedwater temperature	to 230-500°F
Heat transfer surface	capability for testing water walls, preheat, evaporation and superheat tube bundles

The location of the plant is an important consideration, since large quantities of coal and water are required and large quantities of steam are produced. Several advantages could be achieved by locating the development plant on an existing power plant site:

1. Availability of coal-handling, water preparation, and solids disposal facilities.
2. Existing plant would dispose of steam.
3. Superheat and reheat steam generation can be studied by using bleed stream from existing plant.
4. Utility partnership would minimize site development and development time, and provide plant utilities and maintenance facilities.

Thus, the preferred location option would be adjacent to a large power plant where appropriate tie-ins could be affected for supply of utilities, coal, boiler feed water, saturated steam, and return of superheated steam. Such a location would simplify and reduce the cost of installation of the developmental test boiler.

If it is not possible to locate adjacent to a utility, then it will be necessary to install water purification equipment, as well as boiler feed water storage and condensers for steam and adequate boiler feed water pumps. In addition, various auxiliaries such as instrument air, cooling towers, and a packaged boiler would be required at an independent site.

Flow Diagram and Material Balance

Figure 3 is a flow diagram of the pressurized fluid-bed combustion boiler development plant. The plant is adjacent to a large power plant to which it has access to provide interfaces with the coal, water, steam, waste stone, and utilities.

The material balance and operating conditions for the development plant at the projected 100 percent load design conditions are presented in Table 4.

Boiler Module Design

The pressurized fluidized-bed boiler for the development plant comprises a single bed of the four required for an 80-MW module. It is a water-walled box formed of vertical 1-3/4-in. tubing within a 15-ft diameter pressure shell. The box has a rectangular cross-section 5 by 7 ft and a height of 52-feet (Figure 4). The air distributor plate at the bottom of the box can be raised by as much as 4 feet to vary the distance between the base of the bed and the bottom of the horizontal steam tubes submerged in the bed. These steam tubes form a set of 40 parallel vertical platens of serpentine bends filling the cross section of the bed. Three sets of platens are provided so that three different bed heights (and three different amounts of heat transfer surface in the bed) can be provided.

The use of a refractory backed by structural steel for one or more walls of the boiler has been considered as a means of simplifying construction and of easing maintenance. The water wall construction, however, seems to have all the advantages:

Table 4. MATERIAL BALANCES FOR PRESSURIZED FLUIDIZED-BED BOILER DEVELOPMENT PLANT

Stream no.	Description	Temp., °F	Pressure, psia	Flow rate lb/hr	Flow rate 10^6 scfd	Composition,		Particle size
1	Wet coal from BL	Ambient	15	120,000[a]		Moisture VCM Fixed carbon Ash Total	10.0 wt % 36.8 45.3 7.9 100.0 wt %	1.5 in. x 0
2	Sized coal	100	15	111,240[a]		Carbon Hydrogen Oxygen Nitrogen Sulfur Ash Total	71.3 wt % 5.4 9.3 1.3 4.3 8.5 100.0 wt %	0.25 in. x 0
3	Dolomite	Ambient	15	150,000[a]		$CaCO_3$ $MgCO_3$ Inerts Total	49.59 wt % 49.36 1.05 100.00 wt %	0.25 in. x 28 mesh
4	Dolomite	Ambient	15	35,728		Same as stream 3		0.25 in. x 28 mesh
5	Coal to R-101	Ambient	176.4 min.	22,000		Same as streams 2 and 3		0.25 in. x 0
6	Spent dolomite	1,600	up to 330	20,979		$CaSO_4$ CaO MgO Inerts Ash Total	18.0 wt % 39.3 39.6 1.8 1.3 100.0 wt %	0.25 in. x 0
7	Fines discarded	1,600	up to 335	2,900		Ash Carbon Dolomite Total	60.0 wt % 30.0 10.0 100.0 wt %	
8	Fines discarded	1,600	up to 330	1,432		Same as stream 7		
9	Fines discarded	1,600	up to 330			Same as stream 7		

Table 4 (continued). MATERIAL BALANCES FOR PRESSURIZED FLUIDIZED-BED BOILER DEVELOPMENT PLANT

Stream no.	Description	Temp., °F	Pressure, psia	Flow rate lb/hr	Flow rate 10^6 scfd	Composition,	Particle size
10	Air to C-101	Ambient	14.3	366,504	116.8	O_2 20.6 mole % N_2 77.3 H_2O 2.1 Total 100.0 mole %	
11	Air to C-103	100	up to 330	53,500	17.0	O_2 21.0 mole % N_2 79.0 H_2O nil Total 100.0 mole %	
12	Air to R-101	500	up to 335	238,988	76.17	Same as stream 10	
13	Air by-passed	500	up to 335	73,611	23.46	Same as stream 10	
14	Flue gas from R-101	1,600	up to 330	273,737	82.8	CO_2 18.3 mole % CO 0.2 mole % H_2O 8.5 mole % O_2 1.7 mole % N_2 71.2 mole % SO_2 148 ppm NO 198 ppm	20.8 gr/acf
15	Flue gas from C-101	1,600	up to 330	273,737	82.8	Same as stream 14	2.36 gr/acf
16	Flue gas to C-104	1,600	up to 165	60,000	18.1	Same as stream 14	~0.35 gr/acf
17	Flue gas to turbine cascade	1,600	up to 330	213,737	64.7	Same as stream 14	~0.35 gr/acf
18	Quench water	200	up to 330	65,683	131 gal/min		
19	Flue gas to SL-101	700	atm	339,420	116.1	CO_2 11.5 mole % CO 0.12 mole % H_2O 42.9 mole % O_2 1.1 mole % N_2 44.4 mole % SO_2 92 ppm NO 124 ppm	~0.35 gr/acf

[a] Flow rate based on 40-hr operation per week.

1. Lower cost.
2. Lesser bulk.
3. Cheaper fabrication, less maintenance.
4. Better means of support for submerged tubes.
5. More heat exchange surface.

The possibility of locating headers for the tube platens within the water walls has also been considered as a means of easing the removal and/or replacement of tube bundles. More expensive materials and costlier fabrication techniques would be required, but significant savings in carrying out repairs would be realized.

Auxiliary Equipment

Dolomite and Coal Preparation and Feed

Wet coal is received in open-rail cars and in the size range of 1-1/2-in. x 0. The cars are unloaded over a hopper below the tracks. The hopper feeds a conveyor to a dryer where coal is dried from as high as 10 to 3 percent moisture. The coal is crushed to 1/4-in. x 0 and then conveyed to a covered silo for storage. Coal may then be blended with dry, crushed dolomite in the order of 10 percent of the coal. The coal plus dolomite is pressured up to boiler fluidized-bed pressure; then it is injected into the boiler through a Petrocarb pneumatic injection system.

The dolomite is received dry and double-screened 1/4-in. to 28 mesh in covered rail cars. The cars are unloaded into an underground hopper, in a closed building to prevent pickup of moisture. The dolomite is elevated by conveyor to a storage silo. From the storage silo, dolomite is either pressurized in a lock hopper and then injected into the boiler, or dolomite may be added to the coal for injection into the boiler. The dolomite can be injected above the fluidized bed or it can also be added at the bottom of the fluidized bed above the grid.

Spent sorbent is discharged from the bottom of the bed, a position just above the grid or it may be withdrawn at 24, 48 and 72 inches from the bottom of the tube bundle, depending upon the position of the grid with respect to the tube bundle.

Water and Steam Supply

The water and steam are assumed to be available at 2400 psig at the capacity required by the development plant. The water at 500°F and under pressure would be received from the utility power company's economizer and sent to the water walls for preheat and evaporation. Saturated water and saturated steam from the flash drum are returned to the utility company. Either water or saturated steam is taken from the utility company and sent to the fluidized-bed tube bundle. The saturated steam is superheated to 1000°F, or the water is evaporated in the tube bundle and returned to the utility company.

Dolomite and Ash Disposal

Solids disposal is accomplished by cooling in a water jacketed rotary cooler the ash plus the spent sorbent to 300°F from a temperature of 1750°F, with equipment design adequate for 2000°F. The ash is then conveyed to the utility's ash pile for disposal.

Steam Disposal

Normally, all steam is returned to the utility. In case of an independent site without a nearby utility, the steam from all sources—flash drum, tube bundle, or water jackets—would have to be condensed and the condensate less the blowdown would be returned to the boiler feedwater storage and pumped into both the developmental boiler as well as the package boiler.

Particulate Removal

Hot flue gas at 1600-1750°F and at 155 psig leaves the boiler together with most of the ash and some dolomite, as well as a small amount of unburned carbon. The larger-sized solids are removed from the hot gases in a single-stage cyclone. Solids may be returned to the combustion zone if the carbon loss is high, but it is believed that the combustion zone is large enough to insure adequate residence time for complete reaction of the coal. With complete combustion and negligible solid carbon carry-over, the first-stage cyclone dust will be sent to the cooler through lock hoppers. The flue gas will then be sent to a final cleanup consisting of a centrifugal or tornado-type cyclone. Most of the particles 5 μm and larger are removed. If this equipment should prove unsatisfactory for either gas turbine or pollution control, space is allowed for inclusion of a superior type of removal equipment such as sand filters or ceramic filters.

Gas Turbine Test Equipment

The turbine will consist of a stationary test cascade to determine erosion and materials. A full or part size turbine expander will ultimately be used for demonstration purposes.

Combustion Gas Disposal

Flue gases from the test cascade or from the turbo-expander will be quenched to drop the temperture. The pressure will be lowered to atmospheric and then the gas will be sent to a stack through a silencer. In a commercial unit, extra heat recovery equipment and recuperators would be installed on the turbine exhaust gas to recover the most heat possible from the flue gas in order to maximize the thermal efficiency.

Instrumentation and Control

Control of the boiler differs slightly from the standard "once-through" boiler practice but is very similar to process industry's practice. In a conventional boiler, variation in the steam side operation affects the radiant combustion zone only very slightly. However, in a fluidized-bed unit, a slight variation in steam flow rate, temperature or heat transfer immediately affects the fluidized-bed temperature.

For the development boiler, the steam pressure is controlled at 2400 psig, which is in line with modern power plant practice for sub-critical installations. The superheat outlet temperature is limited to 1000°F; this temperature readjusts the water-flow control rate to the tubes in the fluidized bed. The fluidized bed is maintained at the proper pressure by back-pressure control on the effluent gas or on a bypass around the turbo-expander. The temperature in the fluidized bed is used to automatically readjust the coal feed rate after which the air rate is manually adjusted to provide the excess combustion air at the desired percentage of the flue gas.

The reason for manual readjustment of the air rate is that a very slow response is desired so as not to destroy the fluidized-bed characteristics of the boiler. Too rapid a response in air rate may either entrain the entire bed or cause the fluidized bed to collapse.

The emergency control systems will involve immediate stopping of the flow of coal to the boiler. This will occur under such conditions as:

1. Power loss or failure.
2. Instrument air loss.
3. Low pressure in fluidized bed.
4. High pressure in fluidized bed.
5. Low pressure combustion air.
6. Low pressure injection air.

If the coal flow is halted by an emergency condition, the air flow will continue for five minutes and then slowly bypass the boiler. The air compressors will then be stopped automatically.

The demonstration of satisfactory operation of a fluidized-bed boiler depends upon both the efficient generation of steam as well as upon the reduction of pollutants such as SO_2, NO_x and particulates. To reasonably demonstrate satisfactory pollution control, the effluent flue gas will be monitored for particulates, SO_2, NO_x as well as for the usual effluents of CO_2, O_2, and CO.

Particulates can be monitored by means of a beta gage attenuation of particles deposited on a moving tape (Freeman Laboratories) or by means of particles deposited on the surface of piezoelectric quartz crystals (Termal Systems, Inc.). These methods allow a calculated value of particulate concentration in weight of solids per unit volume of gas flow; but it is also necessary to determine particle size distribution for proper operation of cyclones, dust removal equipment, and for extended operation of the turbo-expander. The size distribution can be determined intermittently by means of multi-jet impactor (Monsanto Enviro-Chem Systems, Inc.) or by a stack gas sampler (Anderson 2000).

Flue gas components such as SO_2, NO_2, CO_2 and SO_3 can be analyzed by infrared and/or ultraviolet methods; oxygen by paramagnetic methods; and NO_x by chemiluminescence.

Regeneration/Sulfur Recovery

The calcium sulfate produced in the boiler can either be disposed of as a solid or it can be processed to regenerate $CaCO_3$ or CaO sorbent and to recover sulfur. A number of processes have been proposed for regenerating a sulfated limestone or dolomite. Two processes appear most attractive in conjunction with fluid-bed combustion boilers.

1. Reduction of calcium sulfate to calcium sulfide with H_2 and CO_2 and subsequent regeneration with steam and CO_2 —reaction temperatures below 1600°F at elevated pressures.

2. Direct reduction of calcium sulfate to calcium oxide and SO_2 with H_2 and CO—reaction temperatures greater than 2000°F at elevated pressure or near 2000°F at atmospheric pressure.

Four approaches to regeneration/sulfur recovery have been considered for the development plant:

1. Integral boiler—regeneration—(sulfur recovery) system
2. Two partially independent systems—boiler and regenerator/(sulfur recovery).
3. Regeneration system with flexibility to evaluate alternative regeneration/sulfur recovery processes.
4. Once-through operation.

Unlike the boiler where a design concept is established, sufficient data are not available to permit the selection of a preferred regeneration process. A process may be selected for the development plant on the basis of available data. However, the available data are inadequate to design equipment. Several key parameters which determine the equipment sizing are unable to be specified. Areas of uncertainty include:

1. Sulfur in the spent stone and stone circulation rate from the boiler.
2. Regenerator operating conditions which determine the quantity of heat which must be supplied to or removed from the process.
3. Sulfur dioxide or H_2S concentration which is related to gas supply requirements to the regenerator and the sulfur recovery equipment.

An evaluation was made to determine if a regeneration system could be designed with sufficient flexibility to study alternative processes. Such flexibility cannot be economically provided at this time. Since sufficient information is not available, the plant is designed for once-through operation, and a regeneration system is not included in the preliminary design. Provisions are made for a regeneration system—space, stone feed to the boiler, and removal. Sufficient time will be available to add a regeneration system: the detail design is not scheduled to begin until January 1974; the initial operation of the plant will focus on operation of the boiler plant. The once-through alternative where stone is not regenerated must be weighed with the energy requirements, economics, make-up stone requirements, and environmental parameters of a regenerative system. A once-through system may be the most economic.

Development Plant—Perspective View

A preliminary arrangement of the development plant equipment is shown in the perspective drawing (Figure 5). The fluidized-bed boiler is contained within the vertical cylindrical vessel in the foreground; behind it and to the right is the coal drying and sizing equipment. Coal and dolomite storage silos and feed systems are shown behind and to the left of the boiler. A compressor shelter in the right foreground has been provided for the air supply system.

The entire installation is contained within a 185-x 200-ft site. The top of the pressurized fluidized-bed boiler is about 95 feet above grade; the highest point of the particulate removal system is about 115 feet above grade.

The structure has been designed to provide ease of access to the boiler and its accessory equipment for maintenance and for normal operating checks. Also, gravity flow of solids into the solids feeding vessels from elevated surge bins is utilized to provide maximum solids flow reliability.

EXPERIMENTAL PROGRAM

The experimental program for the pressurized fluidized-bed boiler development plant has three objectives:

1. To verify that the technical findings made with smaller scale equipment (such as those relating to combustion, pollution abatement, heat transfer, materials, etc.) apply directly or can readily be extrapolated to large scale equipment.

2. To study technical problems that can only be studied on larger scale equipment (such as those relating to coal and lime feeding, non-uniform temperature distributions, erosion-corrosion- deposition on moving blades of turbines, etc.) and to demonstrate that no new technical problems (such as vibration and fatigue of boiler tube bundles, etc.) are encountered.

3. To explore advanced fluidized-bed boiler concepts (such as steam generation conditions of 4500 psi/1200°F/1200°F with gas turbine inlet temperatures of 1900°F-2200°F, recirculating bed boilers, etc.).

The end goal is to provide the technical and economic information and to create the confidence necessary for proceeding with the installation of a demonstration pressurized fluidized-bed combustion power plant.

In order to meet these objectives and to reach the goal, measurements are required over a variety of operating conditions—measurements of inlet and outlet flows, compositions, and temperatures sufficient to carry out complete material and heat balances. Such balances permit the computation of combustion efficiencies and heat transfer rates. Analyses of the combustion gas stream emerging from the boiler are required for:

Primary gaseous components—O_2, CO_2, H O, CO, H_2, N_2, and unburned hydrocarbons.
Pollutant gases—SO_2 and NO_x.
Particulates—ash, attrited sorbent, unburned carbon (both composition and particle size distribution).
Trace contaminants.

Some gas composition profiles across the bed and through the bed and disengaging zone are desirable. Occasional measurements of the composition and particle size of solids at various points in the bed are helpful in analyzing boiler operation. Measurements of boiler tube vibration and fatigue and of tube and turbine blade erosion and corrosion are required to estimate long term durability of boiler and turbine. Operating conditions at which measurements should be obtained include primarily the pressure, temperature, air flow, fuel/air ratio, sorbent/sulfur ratio, and bed height over ranges anticipated in the operation of a commercial plant.

To convince utilities that the concept of pressurized fluidized-bed boiler operation is practical as well as economical, the boiler must produce steam reliably, must be capable of easy turndown to 50 percent of its capacity, and must produce combustion gases sufficiently clean to meet pollution control regulations and to provide reliable, long term turbine blade life.

In order to carry conviction, the experimental program must produce data on boiler dynamic operating characteristics, durability of materials of construction in the boiler and turbine, and on the ability to provide any maintenance necessary to achieve uniform and efficient heat transfer, particulate removal from combustion gases, and solids feeding to and from a pressurized system.

To meet the requirements just outlined, the experimental program must demonstrate continuous, controllable coal and dolomite feeding in a sufficient number of feed lines to give uniform bed operation for at least 60 days without interruption.

Concurrent with the boiler proof run, data on NO_x and SO_2 emission, and on particulate content at the turbo-expander inlet will be acquired.

Following a 60-day proof run, the boiler system will be subject to a series of tests to deliberately experiment with the startup, shutdown, and emergency shutdown system, controls, and operating techniques to determine the best methods for coping with and controlling transient phenomena.

The foregoing experimental program will permit the design and construction of a full scale power producing combined cycle pressurized fluidized-bed boiler.

Table 5. OPERATION OF THE PRESSURIZED FLUIDIZED-BED COMBUSTION BOILER DEVELOPMENT PLANT

Phase I

Boiler operating characteristics
(boiler, test passage, no regeneration)

- Ambient temperature and pressure operation
    - Check out solids feeding and withdrawal
        - fluidization
        - particle carry-over
        - water circuitry/boiler tube configuration
    - Use dolomite only
- Ambient temperature, high pressure
    - Check out solids feeding and withdrawal
        - fluidization
        - particle carry-over
        - water circuitry/boiler tube configuration
    - Use dolomite only
- Startup procedure
    - check out using start-up burner and dolomite
    - w/o coal: ability to reach 700-800°F
        - water circuitry
        - air control
- Operate with low sulfur, non-caking coal
- Operate with low sulfur, caking coal
- Operate with high sulfur, caking coal

Phase II

Long term (60 day) system operation

- Boiler control capabilities
    - turndown, operating ranges
- Boiler/gas turbine expander operation
- Operate with boiler/gas turbine expander/regeneration

Phase III

Concept Alternatives

- Advanced steam conditions
- Boiler tube configuration alternatives
- Higher gas turbine temperature
- Possible expansion of plant to a four bed stacked module
- Recirculating bed concept

The operation of the fluidized-bed combustion boiler development plant will be carried out in three phases outlined in Table 5. A recommended schedule for the overall program is presented in Figure 6. In the first phase of the experimental program, operating procedures will be tested; engineering design and performance data will then be gathered on a variety of coals and limestone or dolomite sorbents. In the second phase, overall systems—boiler, gas turbine, and regenerator—control and operation will be investigated; the long term (60 day) boiler run will be carried out. Finally, in the third phase of the program, modifications will be made in the boiler to study advanced boiler designs and operating conditions. Tests will then be carried out to evaluate the effectiveness of such modifications.

IMPLEMENTATION

The schedule in Figure 6 indicates the basic steps toward a commercial fluidized-bed combustion boiler power system which will reduce capital cost, increase operating efficiency, and reduce pollutant emissions for electric energy generation from fossil fuels. Preliminary plans; a detailed design, procurement, and construction schedule; and an overall plant cost estimate will be completed by the end of 1972. These plans, schedules, and estimates will be used in 1973 to locate a plant site, to develop financial backing, and to form a team to construct, operate, evaluate, and lead in the development plant effort. This team should include EPA (and perhaps also other governmental agencies concerned with fuel utilization and power generation), the electric utility industry, electrical generation equipment manufacturer(s), and steam generation equipment supplier(s). Detail design and construction of the development plant would begin late in 1973 or early in 1974. Information from various laboratory, bench, and pilot operations throughout 1973 and 1974 will be factored in the plant design. Operation of the development plant would begin in mid-1975. Sufficient information will be available from the development plant to begin design of a demonstration fluidized-bed combustion boiler power plant in 1977. This plant will be operational late in 1980.

The benefits of such a plant in economic generation of electrical energy from fossil fuels with minimal pollutant emissions amply justify vigorous pursuit of the recommended program.

ACKNOWLEDGMENT

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BIBLIOGRAPHY

1. Evaluation of the Fluidized Bed Combustion Process. Final Report. Westinghouse Research Laboratories. Pittsburgh, Pa. Prepared for the Office of Research and Monitoring, Environmental Protection Agency, Research Triangle Park, N. C. under Contract Number CPA 70-9 (NTIS, PB-211 494). November 1971.

2. Reduction of Atmospheric Pollution. Final Report. The National Coal Board, London, England. Prepared for the Office of Research and Monitoring, Environmental Protection Agency, Research Triangle Park, N. C. June 1971.

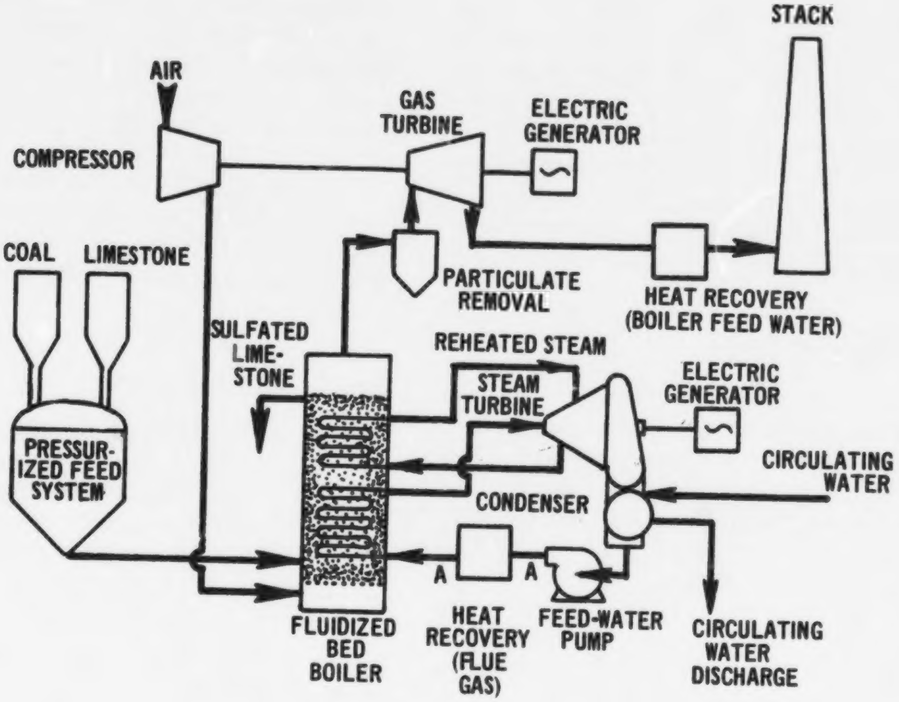

Figure 1. Pressurized fluidized-bed boiler power plant.

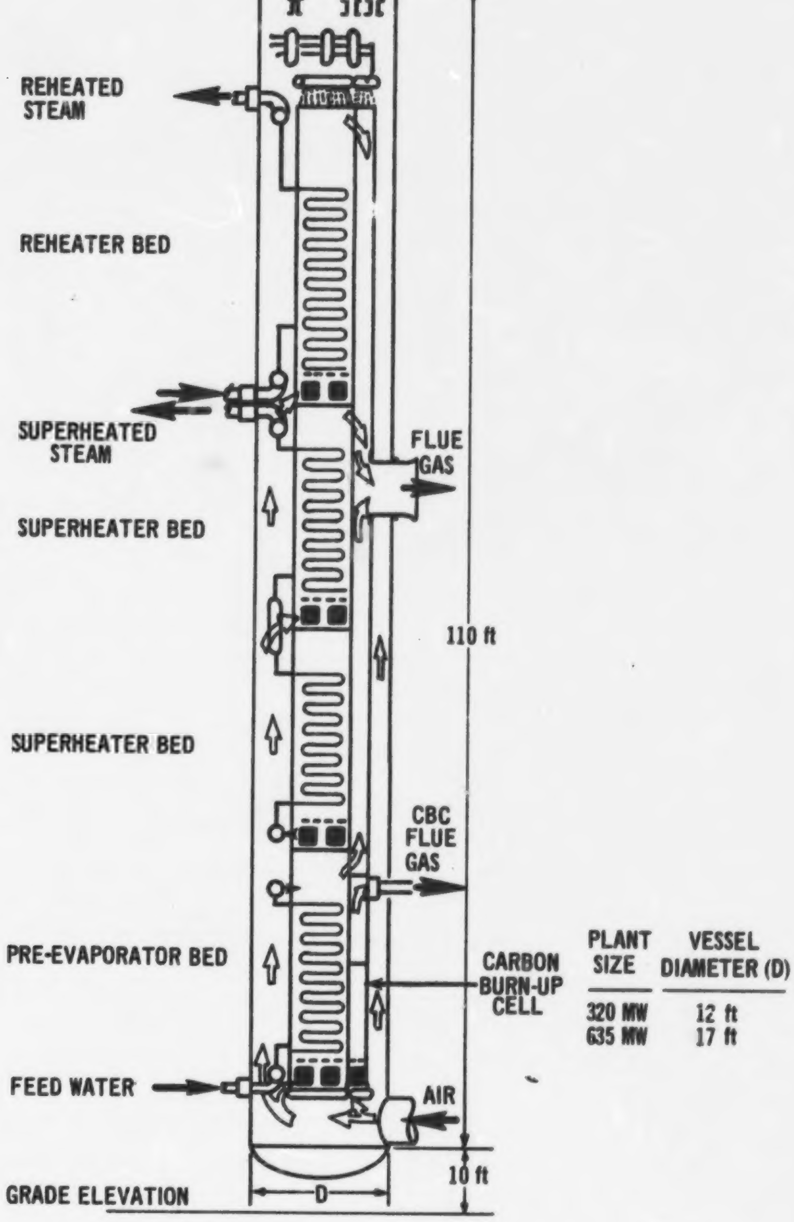

Figure 2. Pressurized fluid-bed steam generator for combined cycle plant (four required).

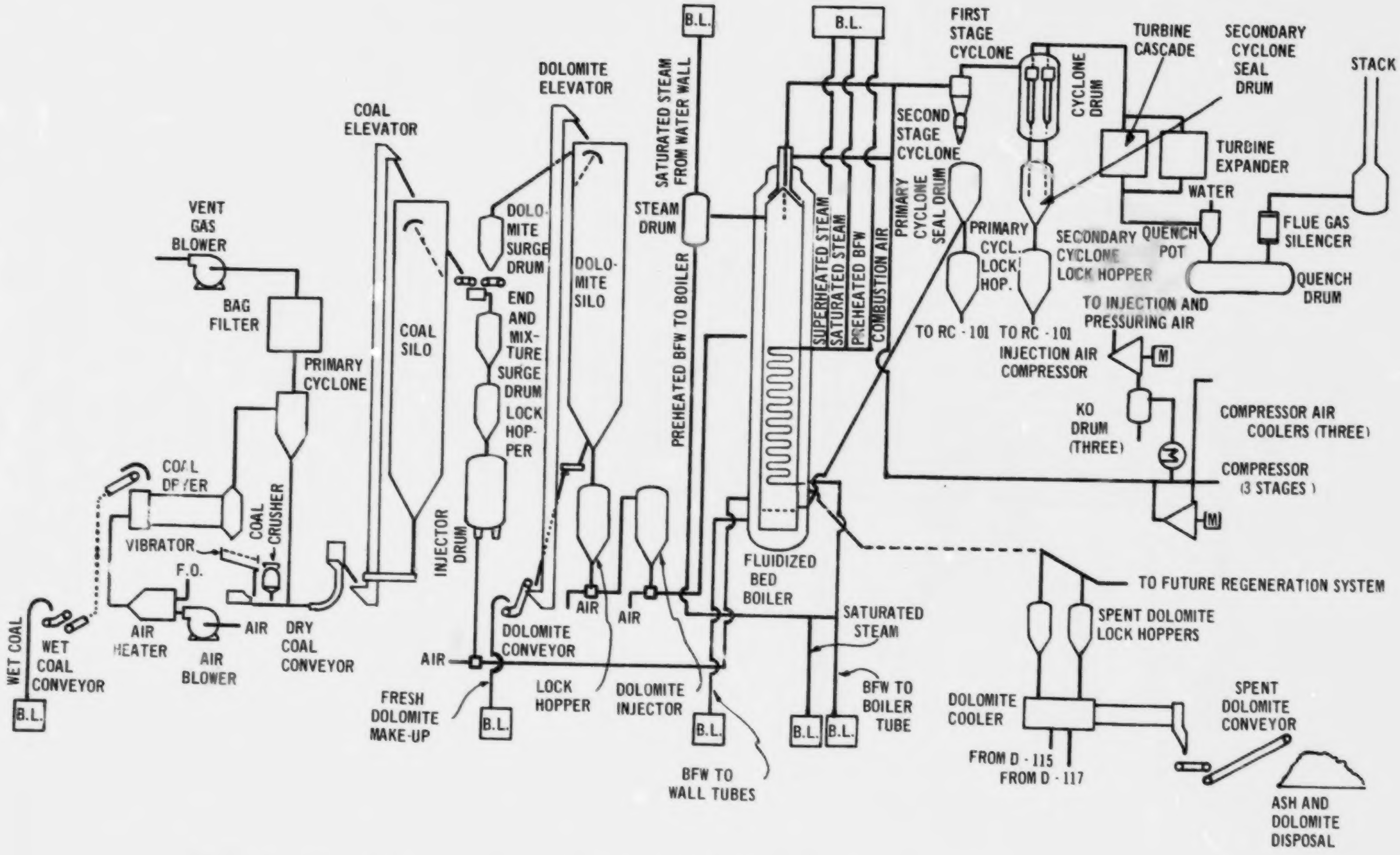

Figure 3. Flow diagram for pressurized fluidized-bed combustion boiler development plant.

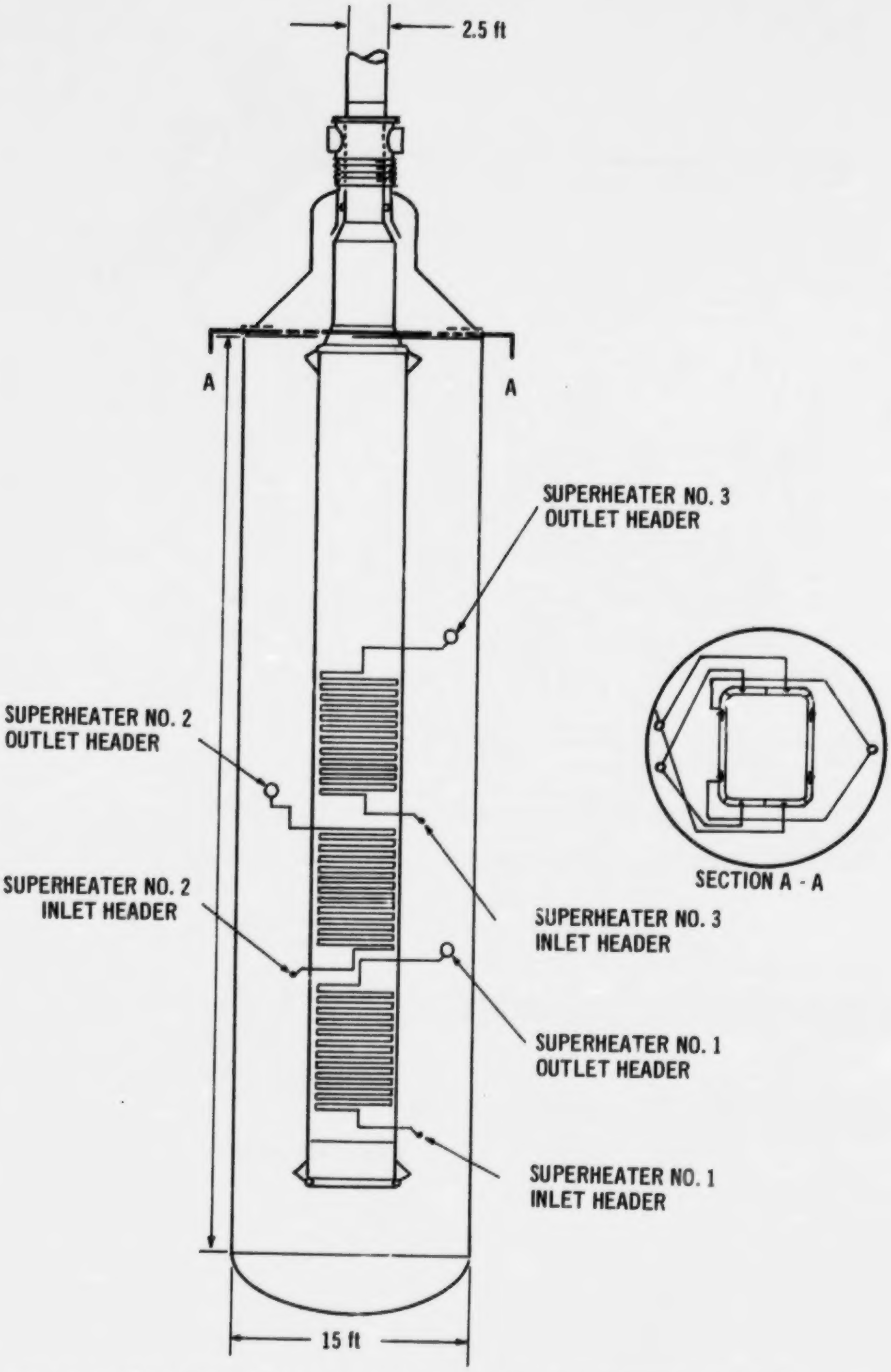

Figure 4. Pressurized fluidized-bed combustion boiler module for the development plant.

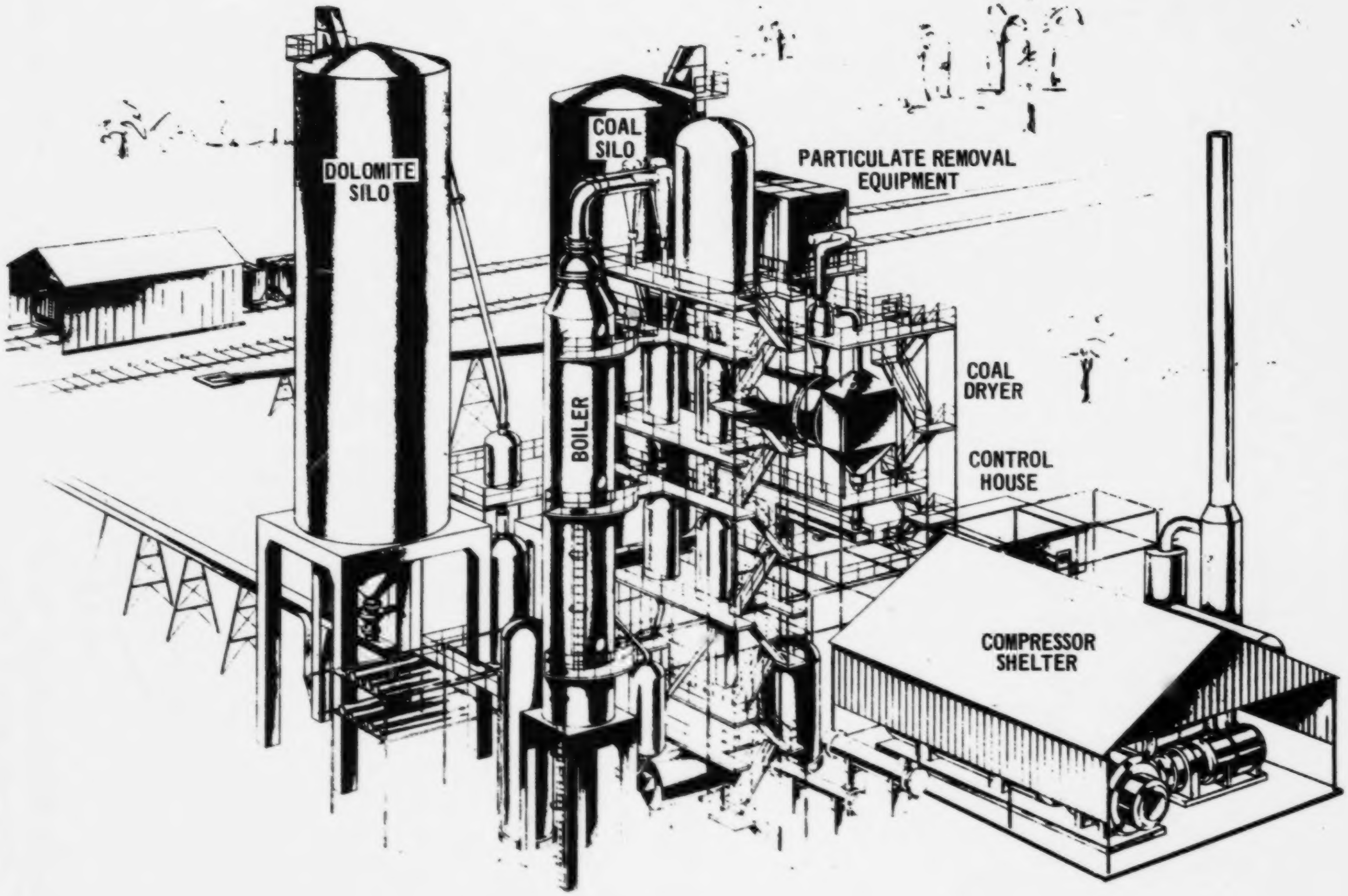

Figure 5. Pressurized fluidized boiler plant perspective view.

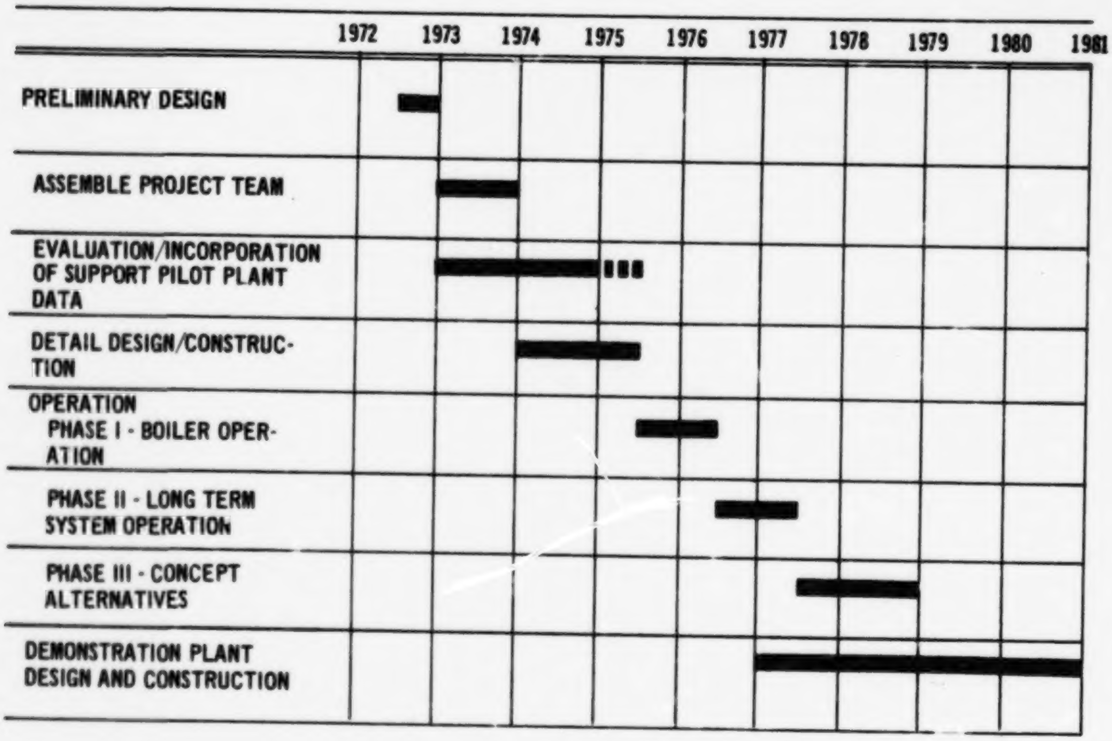

Figure 6. Pressurized fluidized-bed combustion boiler development plant program schedule.

4. GAS TURBINES FOR FLUID-BED BOILER COMBINED CYCLE POWER PLANT

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ABSTRACT

Gas turbines for fluidized-bed combined cycle plants must be engineered to accept hot, high-pressure gases containing erosive dust and small concentrations of compounds that are potentially harmful through hot corrosion and fouling of turbine surfaces. A comprehensive program has been set up to provide gas turbines for these plants. The program involves reducing the concentrations of alkali metal compounds and particulates presented to the turbines; engineering the turbines to accept the hot, high pressure gases; incorporating the turbine design features which make the turbine resistant to attack; and establishing the tolerance of the turbine components for combined hot corrosion-erosion attack.

INTRODUCTION

The fluid-bed boiler combined cycle power plant has the potential of efficiently generating electricity from coal while meeting strict air pollution control standards. Gas turbine expanders are used in such a plant to power the compressors supplying air for combustion and boiler fluidization and to drive alternators supplying about 20 percent of the electrical power output of the plant. In a commercial plant, the hot, high pressure gases leaving the fluid-bed boilers pass through several stages of particulate removal before being admitted to the turbine for expansion. The combustion products at the turbine inlet are anticipated to contain sulfur compounds at the 200 ppm level and particulate loadings in the range 0.1 to 0.2 gr/scf. All of the dust is expected to be less than 10 μm in diameter, and 80 percent is expected to be less than 2 μm in diameter. The erosive potential of these gases should be markedly less than that of the ash resulting from conventional combustion but must be considered in the design of gas turbines to be used with these systems. The off-gases from the fluidized-bed boiler will also contain small, but potentially harmful, concentrations of volatile alkali metal compounds which can react on turbine hardware to produce liquid-films that initiate hot corrosion attack. These condensed liquid films may also catch particulates and cause fouling of the turbine flow passages resulting in loss of turbine performance. The concentrations of condensables entering the turbine are expected to be at least three orders of magnitude lower than would result from conventional combustion of the fuel. This is due to the lower temperatures, longer residence times, and larger surface areas of ash constituents (with which they can react to be retained as solid compounds) in the fluid beds. However, even at small concentrations, attack and fouling of turbine components may eventually be a turbine life-determining factor.

To ensure that a marketable power system results, it is necessary to consider the problem of providing gas turbine equipment able to accept hot, high pressure gases containing erosive dust and small concentrations of compounds that are potentially harmful through hot corrosion and fouling of turbine surfaces. A comprehensive program is in progress to do this.

This effort involves four tasks:

1. Reducing the hot corrosion and fouling potential of the gases leaving the boiler.
2. Design of the turbine to accept the hot, high pressure, particulate containing gas.
3. Design of the turbine to minimize erosion and fouling damage.
4. Determining the hot corrosion-erosion tolerance of the turbine hardware.

The following sections describe the work now in progress in each of these areas.

Reducing the Hot Corrosion and Fouling Potential of the Gases Leaving the Fluid-Bed Boiler

We have begun work to calculate the equilibrium concentrations of condensable sodium and potassium compounds and to estimate the approach to equilibrium that can be expected in the off-gases from the pressurized boilers. This is an extension of previous work done by Boll and Patel[1] (Figure 1)—studying fireside boiler corrosion—to the pressure, temperature, and bed compositions of interest in the fluid-bed combustion system. We are exploring the technical feasibility of using reactions with bed constituents and additives to reduce the concentration of volatiles below those that will allow condensation in the turbine.

Adaption of the Turbine to Accept 1500-1700°F, 10-15 Atmosphere Gases

To preserve high system efficiency it is necessary to transfer the 10 to 15 atm, 1600-1700°F hot gases leaving the dust collection system directly to the gas turbine for expansion. Various design configurations to accommodate thermal expansion, to control leakage, and to provide a uniform distribution of particulates over the flow channel of the turbine have been suggested. Operating experience is available from European compound-cycle power plants utilizing one or two high pressure connections to the turbine. These installations have generally delivered hot gas at turbine inlet temperatures between 1300 and 1400°F, i.e., about 500°F below the turbine inlet temperatures currently used in Westinghouse industrial gas turbines. Gas Turbine Division engineers have prepared a preliminary design using external manifolding to distribute hot gases around the anulus of our W501 (65-MW electric) turbine. This design (Figures 2 and 3) had the objective of avoiding distortion of the turbine casing by non-uniform temperature distributions which they feared would be associated with a single hot gas distributor. Our design engineers are assessing the technical problems associated with these designs, improving them and developing reliability and economic estimates of alternative constructions.

Turbine Design to Minimize Erosion and Fouling Damage

Past experience[2,3,4] with gas turbines expanding dust containing gases indicate design modifications that are helpful in avoiding life-limiting erosion of turbine hardware. Of special concern are design features in the turbine which may cause localized concentrations of the dust in regions susceptible to erosion attack. A turbine design is needed that provides for:

1. Uniform distribution of the dust laden gas over the inlet flow channel.
2. Directs secondary flows in blade and vane wakes to avoid raising the erosion potential of dust at blade and vane roots.
3. Uses stepped side walls, carbide wear resisting inserts, and/or cooling air injection as appropriate to protect blade and vane roots from erosion damage.
4. Appropriately thickens and hard faces blade tips to resist erosion damage.
5. Incorporates spray systems and drains and provides for injection and removal of milled nut shells, washing and cleaning of blade and vane surfaces without the need to open the turbine.
6. Lowers velocity of gases in the turbine, if required, to achieve satisfactory erosion life.

We are studying the effectiveness and the performance tradeoffs of the possible modifications.

Establishing the Erosion and Corrosion Tolerance of Turbine Hardware

We are now developing the experimental design to establish the combined hot-corrosion-erosion damage rates that can be expected on our turbine components. The experimental design involves careful consideration of where particulates will impact, the damage that can be expected from each impact, the interactions between hot corrosion inducing contaminants and erosive particles.

The aerodynamic design of these tests will extend the work of Martlew[5] to the conditions of our turbines and to include the interactions between the cooling air flows, the expanding gas, and the particulates. Martlew's conclusion that only a small fraction of the very fine particles will impact the turbine blades, reinforces the importance of removal of large particulates (Figure 4). The work of Smeltzer, et al.[6] indicates that, if our turbine materials erode in the manner of ductile materials, cumulative damage criteria can be developed that will allow us to predict erosion damage for any distribution of particulates delivered by the particulate collectors.

Establishing the interaction between hot corrosion inducing contaminants and the erosive attack that can be expected is a difficult but necessary task. Fortunately there has been significant recent progress toward understanding the factors involved in the development and retention of protective scales on superalloys[7,8] and on the mechanisms of hot corrosion attack.[9-12] Although many detailed points remain to be resolved, this understanding is optimistically sufficient to allow an experimental design that will provide the data required to engineer long-lived turbines.

We have a comprehensive program underway to provide gas turbines designed to operate on the gases from advanced power plants. The program includes work to clean the gases of trace contaminants. The program is being carried out in cooperation with the Office of Coal Research and the Environmental Protection Agency. Mr. N. P. Cochran, Chief of Utilization, and Mr. W. Moore are acting for OCR. Mr. P. P. Turner, Office of Research & Monitoring acts for EPA. Westinghouse personnel include Mr. G. S. Howard, Manager of Advanced Development, The Gas Turbine Systems Division; Mr. Sven Jansson, Dr. L. Yannopoulous, and Dr. Earl Gulbranson, Inorganic and Physical Chemistry R&D; Mr. Jack Clark, Dr. F. J. Harvey, Dr. C. J. Spengler, and Dr. S. C. Singal, Research Metallurgy. Dr. D. H. Archer, Manager, Chemical Engineering Research is supervising the overall development program.

REFERENCES

1. Boll, R. H. and H. C. Patel. The Role of Chemical Thermodynamics in Analyzing Gas Side Problems in Boilers. J. of Engineering for Power, Transactions ASME. October 1961, pp. 451-467.
2. Smith, J., R. W. Cargill, D. C. Strimbeck, W. M. Nabors, and J. P. McGee. Bureau of Mines Coal-Fired Gas Turbine Research Project - Test of New Turbine Blade Design. Bureau of Mines, U.S. Department of the Interior, Washington, D. C. Report of Investigations Number RI 6920. 1967,
3. Stettenbenz, L. M. Minimizing Erosion and Afterburn in the Power Recovery Gas Turbine. Oil and Gas Journal. 68:65-70, 1970.
4. Atkin, M. L. Australian Coal-Burning Unit. Gas Turbine International. September-October 1969, pp. 32-36
5. Martlew, D. L. The Distribution of Impacted Particles of Various Sizes on the Blades of a Turbine Cascade. In: Proceedings of the Conference of the British Coal Utilization Research Assoc. E. G. Richardson, (ed.). London, Permagon Press. 1960, pp. 104-111.
6. Smeltzer, C. E., M. E. Gulden, and W. A. Compton. Mechanisms of Metal Removal by Impacting Dust Particles. J. of Basic Engineering, Transactions of the ASME. September 1970, pp. 639-654.

7. Wlodek, S. T. The Oxidation of Rene 41 and Udimet 700. Transactions of the Metallurgical Soc. of AIME. 230:1078-1090, August 1964.

8. Tien, J. K. Morphological Study of the Adherence and Growth of Al_2O_3 on Fe-25 Cr-4Al With and Without Yttria or Scandia. (Presented at the AIME Fall Meeting. Detroit. 1971.)

9. Bornstein. N. S. and M. A. DeCrescente. Final Report of the Investigation of Sulfidation Mechanisms in Nickel Base Super Alloys. United Aircraft Research Laboratories, East Hartford, Conn. NSRDL Report 3051, Contract No. N00600-68-C-0639, April 1969.

10. Bornstein, N. S., M. A. DeCrescente, and H. R. Roth. Effect of Vanadium and Sodium Compounds on Accelerated Oxidation of Nickel Base Alloys. Office of Naval Research Report K910983-2, Contract N000 14-70-C-0234 (NTIS, AD 723207). March 1971.

11. Pettit, F. S. and J. A. Goebel. Phase Equilibria Criteria for Hot Corrosion. (Presented at the Metals Engineering Congress. Cleveland. October 18, 1972.)

12. Seybolt, A. U. Role of Rare Earth Additions in the Phenomenon of Hot Corrosion. (NTIS, AD 700-948.) December 1967.

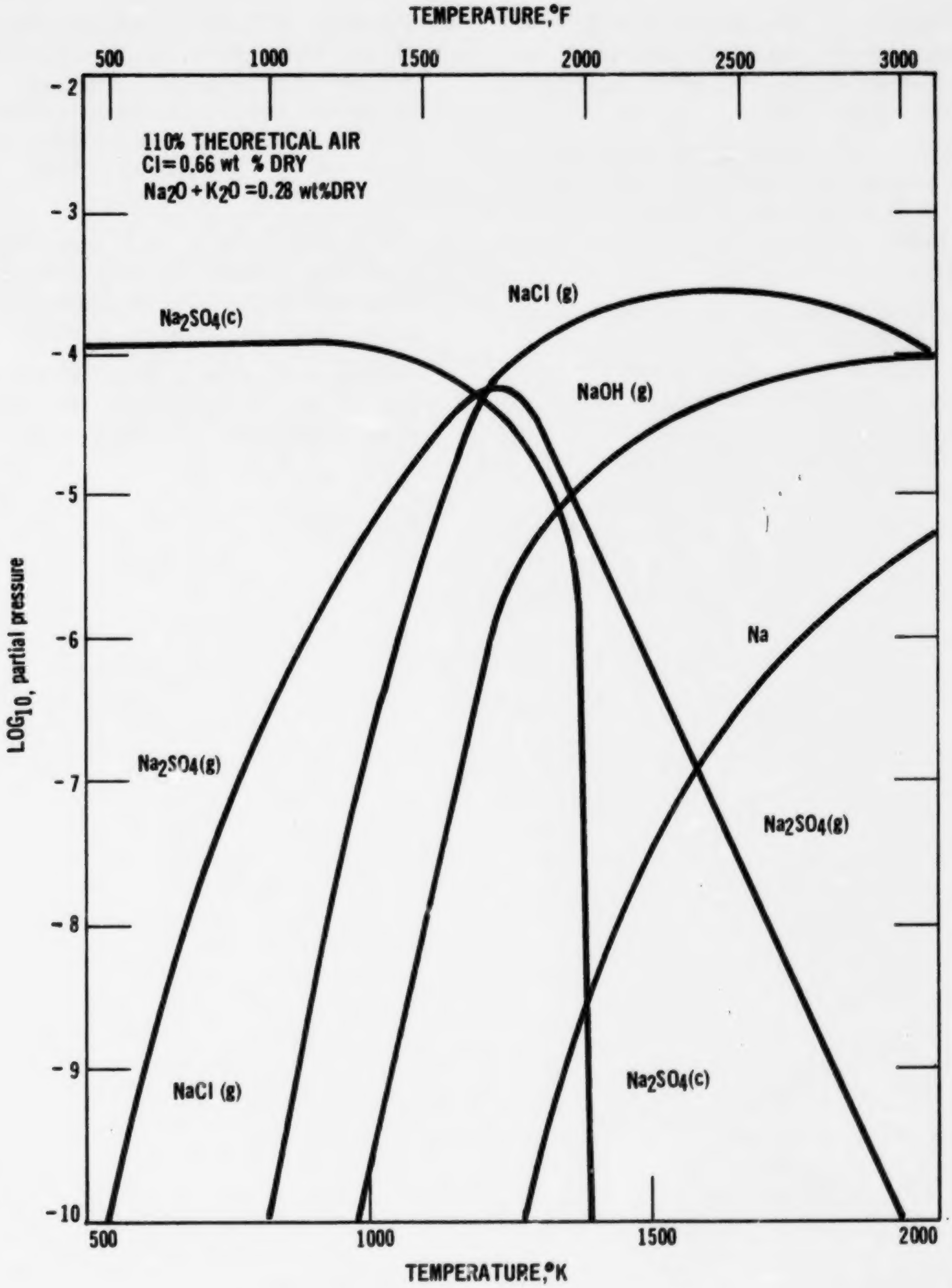


Figure 1. Equilibrium concentrations of alkali metal compounds in combustion of a high chlorine, high alkali coal at 1 atm pressure.[1]

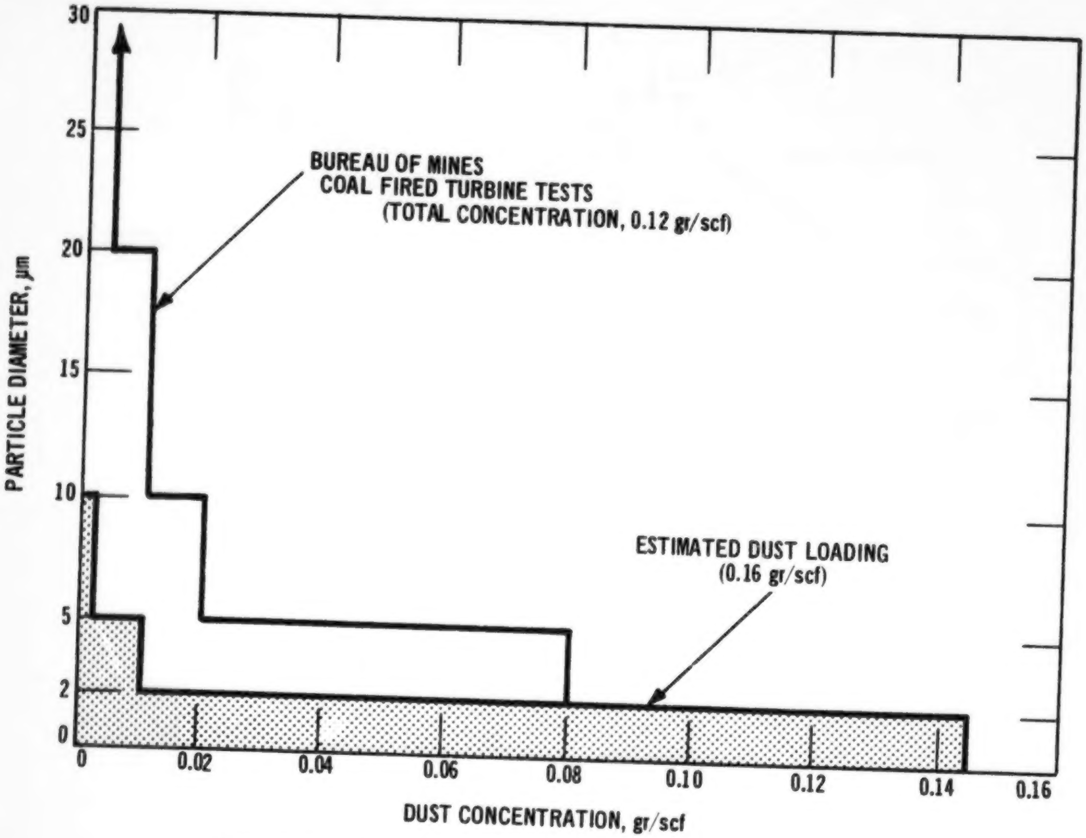
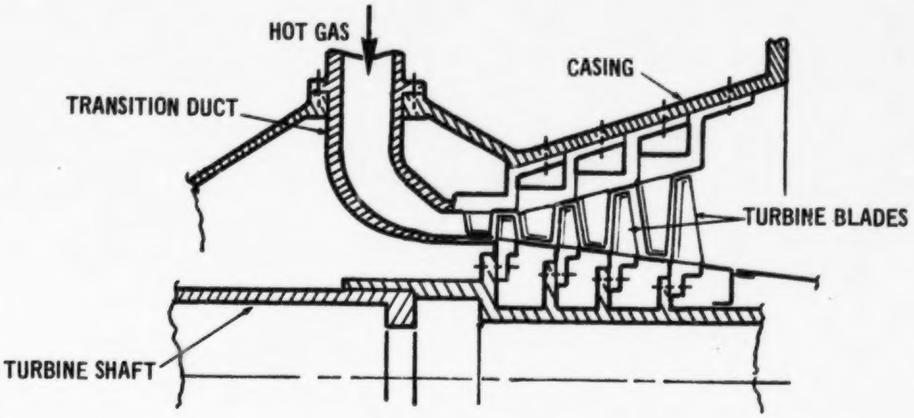
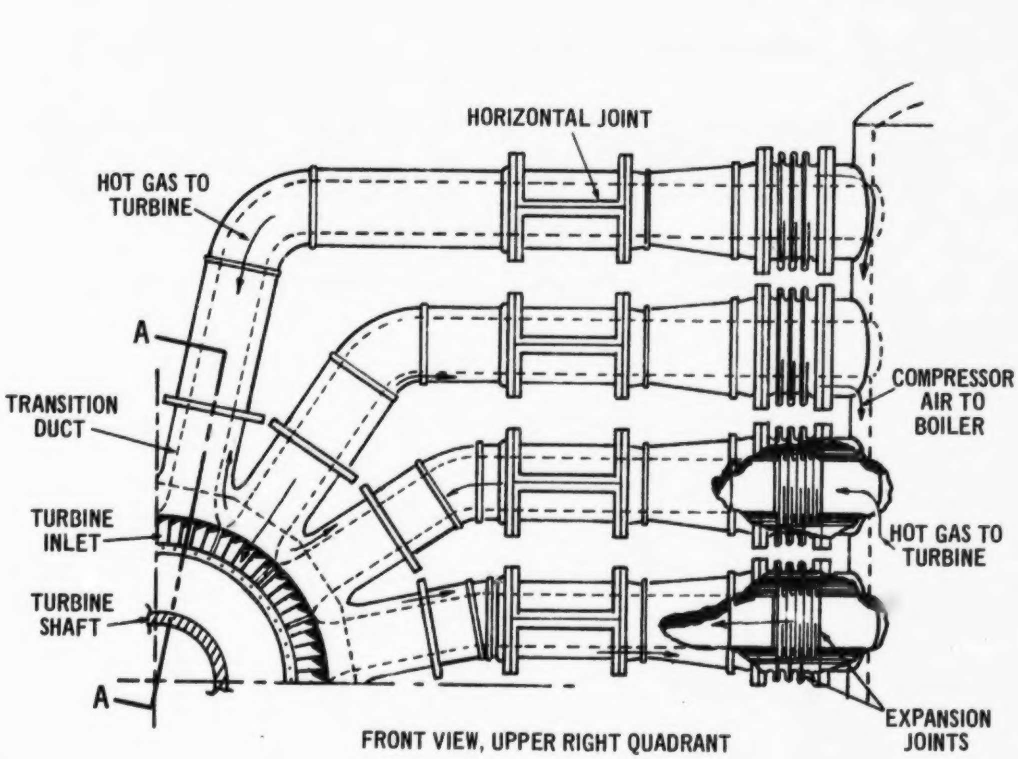


Figure 2. Comparison of anticipated dust concentrations.

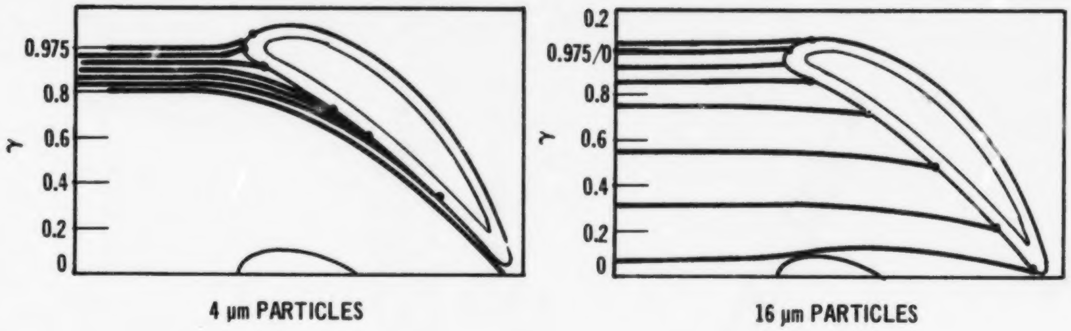


SECTION A-A



FRONT VIEW, UPPER RIGHT QUADRANT

Figure 3. Modification of Westinghouse 501 turbine for hot gas feed.



4 μm PARTICLES

16 μm PARTICLES

Figure 4. Comparison of trajectories of 4-μm and 16-μm diameter particles impacting on a turbine cascade.[5]

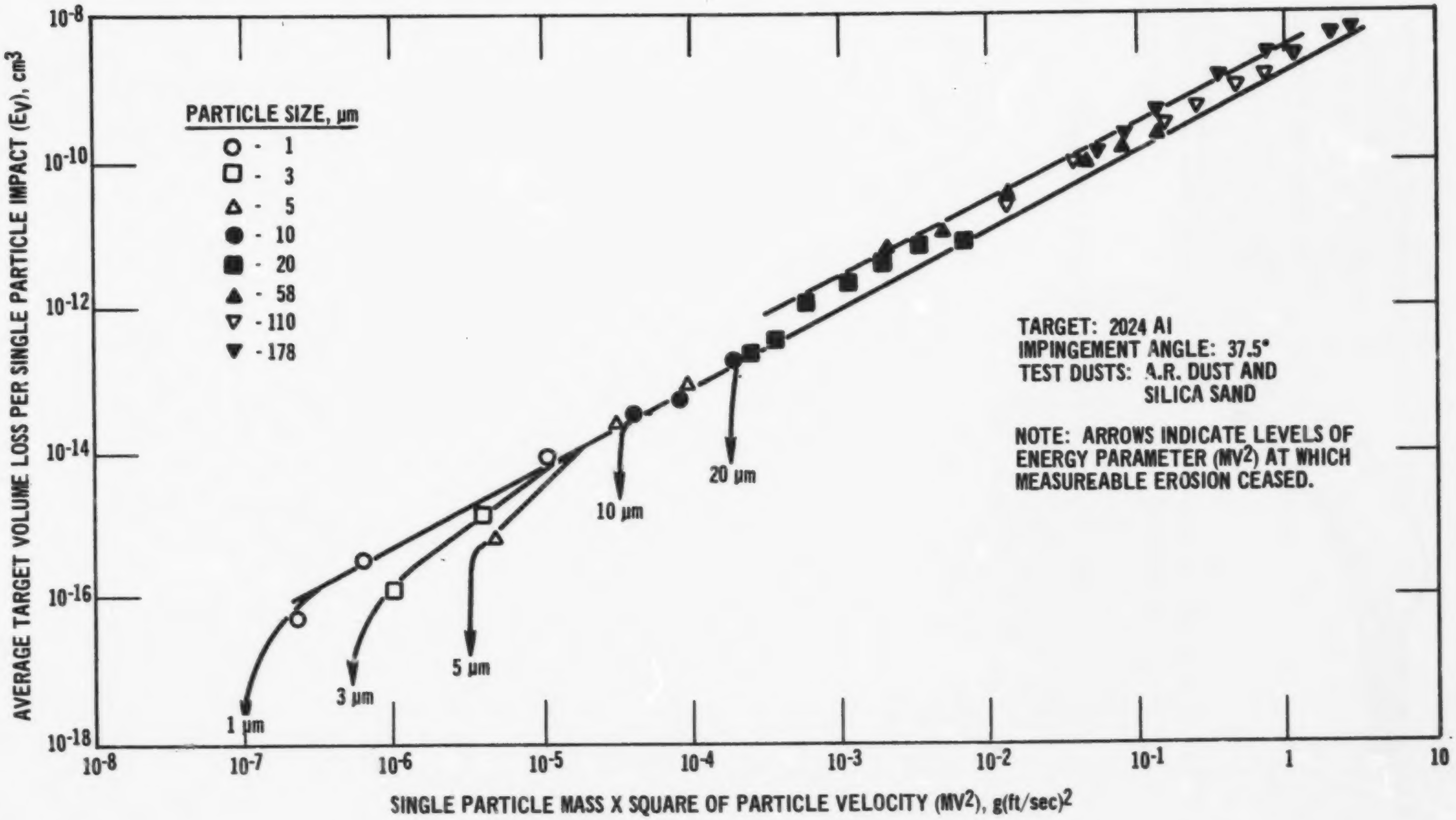


Figure 5. Criteria for estimating cumulative damage from erosion.[6]

5. COMBUSTION OF RESIDUAL FUEL OILS IN FLUIDISED BEDS

H. G. LUNN, A. G. ROBERTS AND H. B. LOCKE

ABSTRACT

The paper gives some insight into the experience gained on the programme of work currently being carried out by BP into fluidised combustion using petroleum-derived fuels. Part of the work is based at BP Research Centre, Sunbury-on-Thames, and part is being carried out for BP by BCURA Ltd. at Leatherhead.

Information is presented on heat transfer, atmospheric pollution, oil injection systems, corrosion of immersed metal surfaces, and the current and projected work programme. High combustion efficiencies (greater than 99.5 percent) are attainable with low NO_x emissions (100 ppm). Sodium and vanadium retentions are encouragingly high; SO_2 emissions can be reduced to low levels by the addition of limestone or dolomite.

INTRODUCTION

Fluid-bed combustion of oil offers the possibility of achieving high combustion intensity with high rates of heat absorption (to tubes in the bed). At the same time operating temperatures are low enough to minimise the release into the gas of fuel constituents; such constituents are known to be deleterious from the point of view of fouling and corrosion of heat transfer surfaces.

Fluid-bed steam generators in comparison with conventional flame systems can be expected to:

1. Be more compact and occupy less height.
2. Require less heat transfer surface.
3. Require less on-site fabrication.
4. Emit less nitrogen oxides to the atmosphere.
5. Be capable of low SO_2 emissions. Depending on the price margins between high and low sulphur fuels, lower operating costs could result.

In addition, the potential for retaining sodium and vanadium in the bed offers the possibility of utilising a supercharged fluid-bed system in combined steam/gas turbine cycles. This would result in still further reductions in size, in capital costs, and in operating costs.

EXPERIMENTAL RIGS

The main experimental equipment consists of two combustors—a 15-in. diameter combustor at the BP Research Centre, Sunbury and a 42-in. diameter combustor at BCURA Ltd., Leatherhead. Both combustors are complementary, with a coordinated development and experimental programme. The 15-in. diameter combustor has burned mainly a light fuel oil (Table 1, No. 1) and the larger combustor either a nominal 3500-sec fuel oil or an atmospheric residue (Table 1, Nos. 2 and 3, respectively). Total operating time is about 100 hours on the 15-in. combustor and 1000 hours on the 42-in. combustor.

Table 1. TYPICAL PROPERTIES OF FUEL OILS USED

	15-in. Combustor	42-in. Combustor	
	1	2	3
Specific gravity (60°F)	0.955	0.955	0.961
Gross calorific value, Btu/lb (approx.)	18,800	18,500	18,170
Total sulphur content, wt%	3.0	2.2 3.0	4.0
Viscosity, redwood no. 1 (at 100°F), sec	1,000	1,500-2,500	3,000
Ash content, wt%	0.01	0.04	0.03
Vanadium content, ppm	45	60 150	50
Sodium content, ppm	40	20 65	26
Nitrogen content, ppm	2,070	2,300 3,000	2,100

Table 2. TYPICAL OPERATING CONDITIONS

	15-inch Combustor	42-inch Combustor
Fluidising velocity, ft/sec	6	6
Bed material	10 x 30 mesh sand	10 x 16 mesh sand
Bed depth, inches	12-20	20-30
Bed temperature, °F	1450-1750	1500-1700
Excess air, %	5-25	5-20
SO_2 acceptor	None	Limestone/Dolomite

Table 3. TYPICAL ANALYSES OF LIMESTONE AND DOLOMITE

Component	Composition, %	
	Limestone 18 [a]	Dolomite 1337 [b]
CaO	45.7	28.9
MgO	1.4	22.9
$H_2O + CO_2$	36.6	47.4
SiO_2	13.6	0.5
Fe_2O_3	0.3	0.2

[a] Limestone 18: Supplied by Fuller Industries Inc., Florida.

[b] Dolomite 1337: Supplied by Charles Pfizer & Co., Ohio

15-inch Diameter Combustor (Figure 1)

The rig is in the form of a 15-in. diameter stainless steel cylinder, made up from 12-inch long flanged sections. The construction allows any part, including the distribution and heat exchange sections, to be removed and replaced with a minimum of time and effort. The whole of the hot portion of the rig is contained in an insulated"cupboard." There is a fixed area of heat exchange surface in the fluid bed which removes about 180,000 Btu/hr as hot water. The flue gases pass through an expanded convection section where a finned tube heat exchanger can remove up to 70 percent of the heat in the flue gas, again as hot water. The rig has fully automated startup and shutdown procedures and can be operated easily by one man.

42-inch Diameter Combustor (Figure 2)

This plant was originally designed as a prototype vertical shell boiler burning coal in a 48-in. diameter fluidised bed; it operated for several hundred hours in this capacity. It was then modified for oil-firing investigations. The fluidised bed is contained in a refractory-lined chamber suspended below the boiler furnace tube. Cooling tubes (1-1/2-in. OD) extend across the bed to remove some of the input enthalpy; the number of tubes in use are altered to produce any desired bed temperature. The freeboard is enclosed by a water/steam jacket, and the sensible heat in the exhaust gases is reduced by heat exchange with the water in a three-pass exchanger. Material elutriated from the boiler is separated partly in a settling (gravity) chamber and subsequently in a cyclone dust collector unit. The plant, including feedwater system and solids addition and disposal, can be easily operated by two men.

Typical operating conditions for both combustors are given in Table 2.

COMBUSTION

Initial experience with the 42-in. combustor showed that burning a heavy residual oil in a fluidised bed was not such a simple matter as might have been expected. The combustion air has to serve two functions—as combustion air and as fluidising medium—but the residence time of the oil in the bed is very short so that rapid mixing of oil and air is vital.

The following principles have been established as desirable if a high combustion efficiency with minimal burning in the freeboard and no smoke emissions are to be achieved.

1. The oil should enter the bed at a large number of points over the cross-section of the bed. Ideally all the oil and combustion air should be mixed before entering the bed. This is a logical consequence of the relatively poor mixing of the bed in a lateral direction and the almost immediate vapourisation of the oil as it enters the bed.
2. The oil should be admitted at a location where the bed is fully fluidised. Otherwise the oil and bed material will form a sticky agglomerate with a tendency to coke and accumulate, which will eventually result in poor fluidisation. This requirement contradicts the previous principle in that part of the air must be used solely as fluidising medium.

There are a number of possible design solutions. One which has been adopted on the experimental rigs is shown in Figure 3. Air is admitted from a plenum chamber through a number of nozzle caps in the distributor plate. These caps communicate with an oil reservoir through small holes; an oil film is carried up the caps by the air flow and injected into the bed. The oil holes act either as restrictors or as weirs (depending on the viscosity and flow rate of the oil) and produce a more or less even flow of oil to each cap. The oil is preheated to about 150°F.

Additional caps (not shown in Figure 3) admit a proportion of the combustion/fluidising air at a level slightly below the oil inlets. Between 50 and 70 percent of the total number of caps are used in this way to fluidise the bed at a level below the oil injection points.

The distributor assembly also contains a propane reservoir from which gas is drawn into the caps by the air flow. In this way a substantially pre-mixed gas/air medium is fed into the fluidising bed with minimal risk of an explosion in the plenum chamber. This facility is used only for startup and for raising the bed temperature to a sufficiently high level for oil to be injected.

For startup, a gas/air burner is ignited in the freeboard just above the bed surface, with the full amount of fluidising/combustion air passing through the bed; at this stage the bed is not fluidised. Propane is then admitted to the distributor system and ignited on the bed surface. The ignition plane then slowly descends through the bed, raising the temperature of the latter, and causing fluidisation in the process. The whole procedure can be speeded up by using an above-normal air flow through the bed initially, and then reducing the flow as the bed temperature rises. The bed temperature is raised to 1300/1500°F in about 15 minutes, at which time the change-over to fuel oil can be carried out.

Using this system (with slight variations between the two rigs), combustion efficiencies are greater than 99.5 percent when burning any of the oils listed in Table 2.

ATMOSPHERIC POLLUTION

SO_2 Emission

The fluidised bed provides a suitable environment for retaining sulphur if a suitable acceptor is fed to the system. Tests have been carried out in the 42-in. combustor with both U.S. limestone 18 and U.S. dolomite 1337 (see Table 3) fed by gravity into the bed. As expected, SO_2 emission can be reduced to almost any desired level by adding sufficient calcium. The quantities required increase with bed temperature, and dolomite is slightly more efficient than limestone (on a Ca mole basis). At a bed temperature of 1550°F, fluidising velocity of 6 ft/sec, bed depth of 2-ft and with limestone 18 (sized 1/8 inch x O) as the acceptor, a sulphur dioxide emission of less than 0.8 $lb/10^6$ Btu heat input* from an oil of 2.7 percent S content requires a Ca/S mole ratio in the feed of 2.5 to 1. This corresponds to a limestone 18 feed rate of 0.26 lb/lb oil. The quantity could be reduced considerably by recycling the material elutriated from the bed.

Tests are currently in progress to elucidate the effect of bed height, excess air and sulphur content of the oil on SO_2 emission. Evidence to date suggests that the effects are the same as for coal firing, and that in practice oil and coal behave in a similar manner.

NO_x emission

This has been recorded consistently at 100 ppm v/v ± 30 (about 0.2 $lb/10^6$ Btu). Three different methods have been used — chemiluminescence, infra-red, and chemical methods. All the methods have given consistent and similar results. The currently-proposed limit for NO_x emission in the U.S.A. is 0.3 $lb/10^6$ Btu. Fluidised combustion of oil would seem to offer the possibility of achieving this limit at no extra cost.

When burning propane, even lower NO_x values of less than 40 ppm are recorded. The actual value, with either oil or gas, seems to be independent of the likely range of operating conditions in a fluidised bed.

Particulate emission

Solids emission is very much a function of plant design (in particular, freeboard height and gas cleaning facilities), and results obtained on the experimental combustors can only be used as a general guide. In the 42-in. combustor when adding limestone into a sand bed at a rate of about 0.2 lb/lb oil, the stack dust emission is 0.15 to 0.20 $lb/10^6$ Btu with a size distribution of 98 percent <30 mesh (500 μm) and 56 percent <200 mesh (76 μm).

HEAT TRANSFER

Heat transfer rates to water-cooled tubes immersed in the bed have been measured in both combustors. The effect of type of bed material on heat transfer was investigated in the 15-in. diameter combustor over a range of size distributions and fluidising velocities using crushed firebrick, tabular alumina, sillimanite, chrome ore, and sand. It was found that, with the exception of sand, the results agreed with data obtained in the U.K. in coal-fired combustors when coal ash was the bed material. With sand the heat transfer rates were 10 to 15 percent higher than with the other materials; the causes are unknown, but could be related to particle shape factor.

Similar results were obtained in the 42-in. combustor. With sillimanite as the bed material, heat transfer rates agreed with predictions from earlier coal-fired tests. With sand, however, heat transfer rates were 10-15 percent higher—a typical bed-to-tube coefficient when using a sand feed of 10 x 16 mesh at a superficial fluidising velocity of 6 ft/sec and a temperature of 1575°F was 60 $Btu/ft^2\text{-}hr\text{-}^\circ F$. This corresponds to a mean bed particle size of 1300 μm.

CORROSION OF TUBES IN THE BED

A number of alloy specimens are immersed in the bed of the 42-in. combustor to assess the possibility of corrosion problems. The specimens are accurately machined in tubular form and are welded together to form loops which are controlled to appropriate metal temperatures by internal air cooling. The alloys being used, and the corresponding metal temperatures are:

2.25 percent chrome (800 - 1050°F)
12 percent chrome (1100 - 1200°F)
AISI 347 Austenitic Steel (1100 - 1250°F)

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*This is a currently proposed limit in the U.S.A.

Metallographic examination revealed no signs of corrosion attack of any of the specimens after a test of 100 hours duration, and weight losses were similar to or less than those published[1,2,3] for conventional oil-fired boilers. More conclusive information will be obtained from a test of 500 hours duration which is currently in progress.

COMBINED CYCLE APPLICATION

Fluidised combustion processes offer a potential reduction in capital costs compared with conventional equipment, and these benefits are increased if the process can be carried out under pressure. There is also a potential reduction in operating costs if the process can be utilised in a combined steam/gas turbine cycle. However, the energy in the combustion products must then be recovered in a gas turbine.

As a first stage in assessing whether the flue gases from an oil-fired fluidised bed can be passed over gas turbine blades, the behaviour of sodium (Na) and vanadium (V) in the experimental rigs is being investigated.

The 15-in. combustor is currently carrying out a long-duration test in which a bed of fresh 10 x 30 mesh sand is being operated for as long as possible without further additions. Samples of bed material are withdrawn at regular intervals for determination of Na and V content. An attempt will be made to determine the maximum amounts of sodium and vanadium that can be retained by the bed material and the form in which they are present.

Measurements of the Na and V present as aerosol in the flue gases from the 42-in. combustor show that about 70 percent of the Na and more than 99 percent of the V is retained in the bed material when burning the oil No. 2 of Table 1. These results are sufficiently encouraging to consider the next stage in the development. It is intended to modify the coal-fired pressurised combustor at Leatherhead (Figure 4) so that it can also be operated with residual fuel oil. This combustor has been described at previous meetings. It is proposed to carry out tests at 5-6 atm pressure and temperatures in the range 1470-1700°F. The products of combustion pass over a static cascade of turbine blades.

REFERENCES

1. Rossborough, D. F. Mitt VGB, No. 51, February 1971, p. 51.
2. Holland, N. H., et al. Journal of the Institute of Fuel. May 1968, p. 206.
3. Jackson, P. Third Liquid Fuels Conference, Institute of Fuel, 1966.

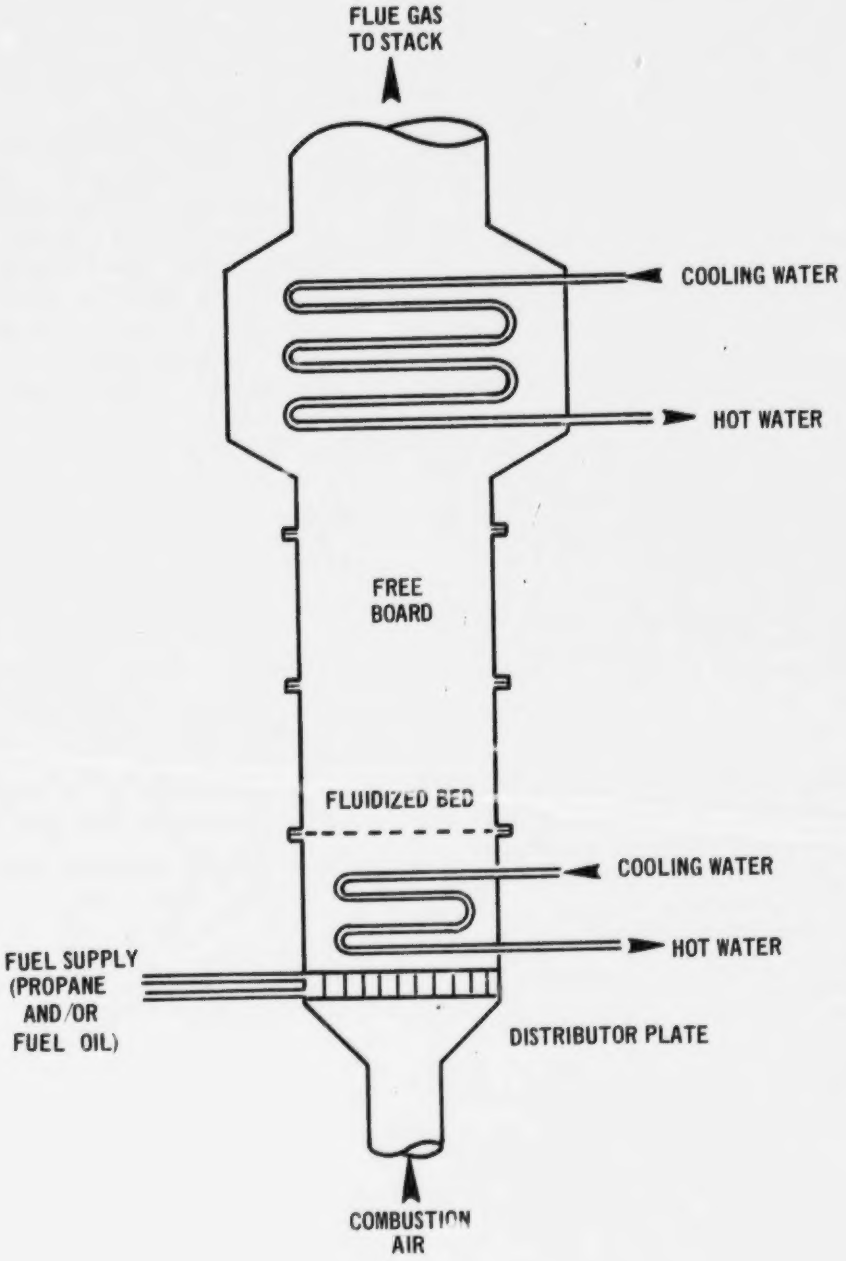

Figure 1. 15-inch diameter combustor.

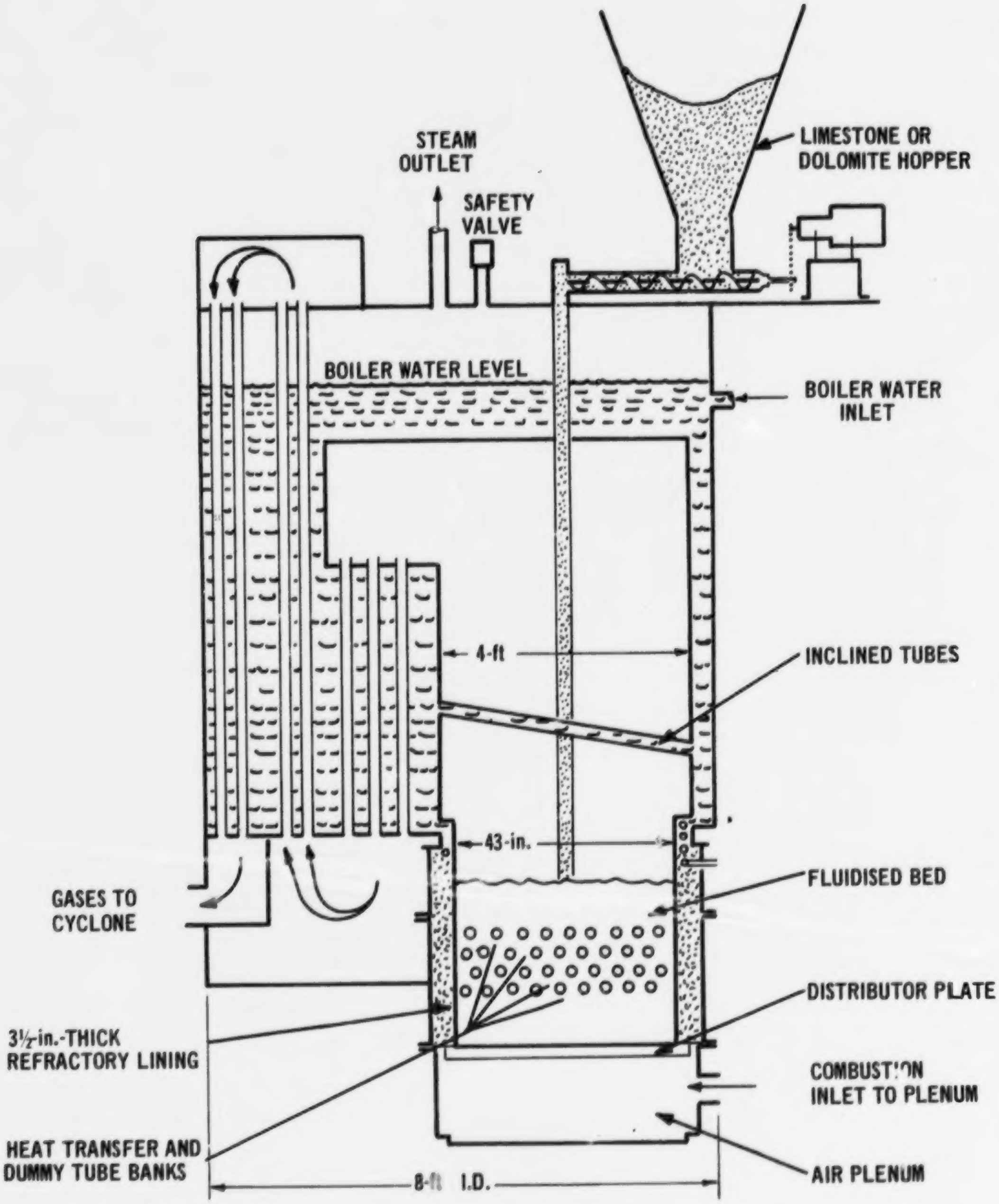

Figure 2. 42-inch-diameter combustor.

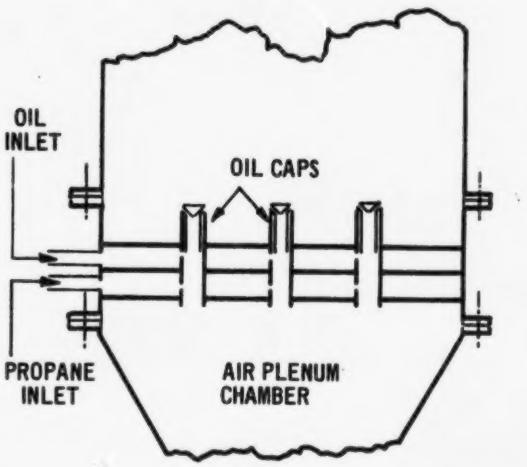

Figure 3. "Climbing film" distributor.
(Patent applied for.)

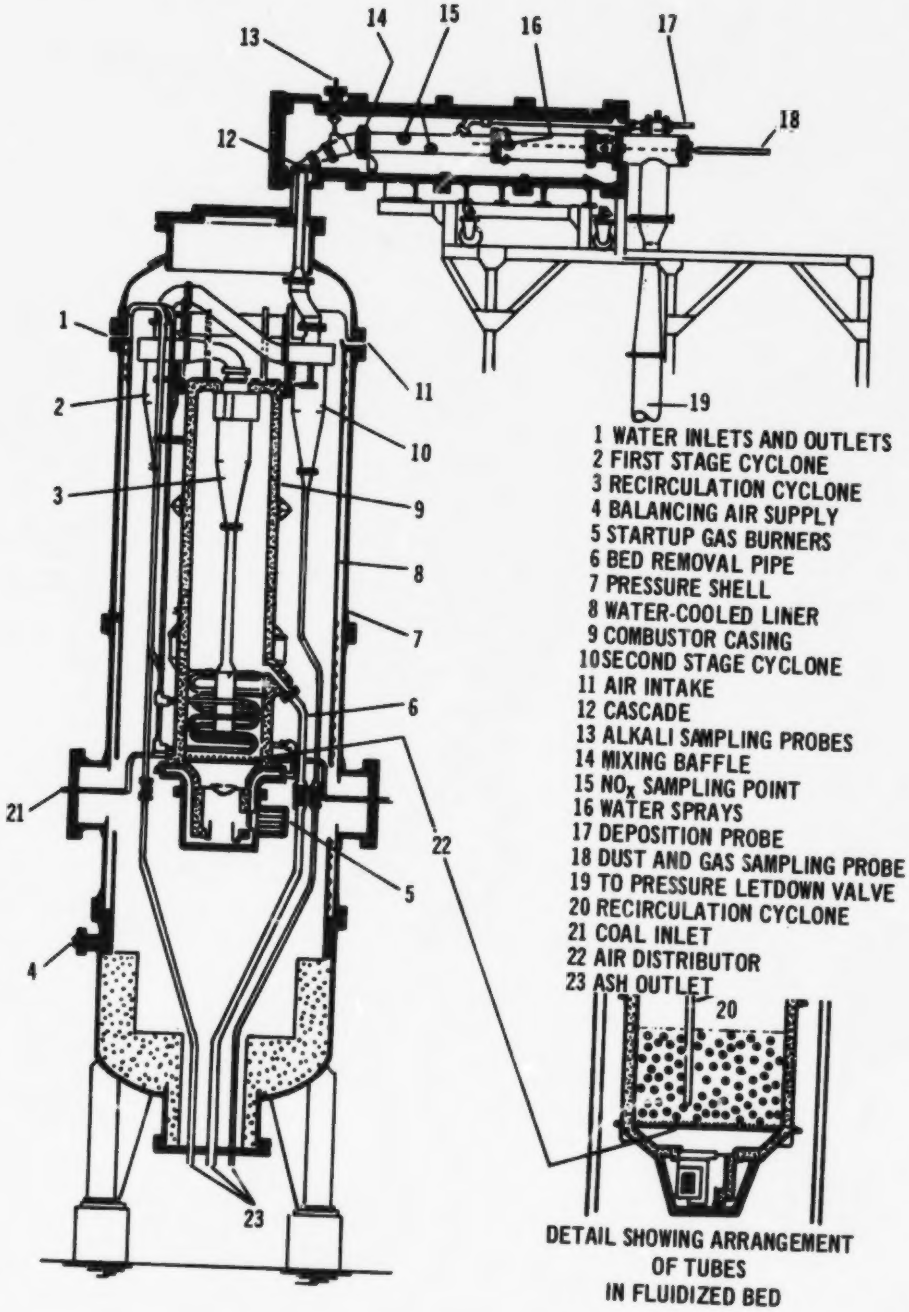

Figure 4. Coal-fired pressurised combustor.

6. FLUIDIZED-BED AIR HEATERS FOR OPEN AND OPEN/CLOSED GAS TURBINE CYCLES

H. HARBOE

Stal-Laval (G. B.) Ltd., U. K.

The heat transfer coefficient between bed and cooling tubes in a fluidized-bed combustor remains nearly constant over the permissible operating regime for a given design.

This is a feature which gives rise to substantial control problems when the combustor is used as a boiler. It is generally accepted that it will be necessary to resort to bed-level control or compartmenting of the bed to obtain satisfactory turndown ratios.

If, instead, the combustor is used as an air (or gas) heater it will be possible to overcome these difficulties and readily achieve controlability from zero to full load.

Water and steam in the cooling tubes will give a nearly constant tube wall temperature much below the bed temperature. With air in the tubes the tube wall temperature can be allowed to increase and thus provide much bigger variations in the difference between bed temperature and tube wall temperature.

As an example, with 108 atm air in the cooling tubes the load can be reduced from 100 to 50 percent by a reduction in bed temperatutre from 850 to 800°C (1562 to 1472°F). If the tubes are cooled with 127 atm steam the corresponding reduction in load would be from 100 to 86 percent.

The air heater alternative carries the penalties of requiring more tube surface and better quality tubes, but preliminary design exercises show that these penalties can be offset against much greater simplicity.

One air heater design concept is for a 360-MW generating unit. This basically comprises the components from an 80-MW intercooled open cycle gas turbine, the compressors of which provide all the combustion air at 18 atm. A 140-MW almost-closed cycle air turbine working between 18 and 108 atm gets its heat input from the cooling tubes in the bed (it is only "almost-cooled" because it is connected on the L.P. side to the H.P. side of the open cycle unit). Cooling between turbine and compressor in the closed cycle unit is done in a simple once-through boiler which together with a waste heat boiler after the open exhaust turbine can raise steam for a 140-MW low pressure steam turbine. The exhaust turbine and the closed cycle unit will together drive a 345-MW main alternator, the open cycle compressors, plus the steam turbine; a 15-MW alternator will be joined along a separate shaft.

Preliminary evaluations show that this arrangement can control the load down to idling by throttling on the fuel and on the inlet to the open compressor—thereby also altering the pressure level of the "closed" cycle—without reducing the bed temperature below 705°C (1300°F).

Layout proposals indicate that the entire 360-MW unit including combustors can be housed in a building having only 40 percent of the volume of a conventional boiler-turbine installation. Detailed costing for this plant is still in progress.

Proposals have also been worked out for conventional open cycle gas turbines using fluidized-bed combustors and of special interest may be the use of fluidized-bed combustors associated with air storage plants.

Fluidized-bed combustion is likely to find its biggest market for midload type of plant and this in turn demands frequent stops and starts and good controlability. Not only can a fluidized-bed air heater be more easily controlled during operation than a fluidized bed boiler, but it is also to be expected that an air heater can more easily be shut down and restarted.

It is suggested that a 3-MW and a 65-MW open cycle gas turbine with fluidized-bed air heaters should form two development stages prior to the 360-MW combined open/closed cycle unit.

SESSION VI:

Discussion Panel and Summary

PANEL MEMBERS:

Mr. R.P. Hangebrauck, EPA, Chairman
Professor D.E. Elliot, University of Aston, England
Mr. D.B. Henschel, EPA
Mr.H.B. Locke, National Research Development
Dr. A.M. Squires, City College of the City University of New York

1. MINUTES OF PANEL DISCUSSION AND SUMMARY SESSION

D. B. HENSCHEL

Environmental Protection Agency

The final session of the Third International Conference on Fluidized-Bed Combustion was a general discussion, led by a panel, covering various topics of importance that had been raised during the Conference.

Members of the discussion panel were Mr. R.P. Hangebrauck (Chairman), Dr. D.H. Archer, Professor D.E. Elliott, Mr. H.B. Locke, Professor A.M. Squires, and Mr. D.B. Henschel.

Mr. Hangebrauck (EPA) opened the discussion by asking the panel members and the other attendees for their conclusions regarding the advantages of regenerating the spent SO_2-control sorbent that would be produced in a fluidized-bed boiler. Dr. Archer (Westinghouse) stated that economics will probably favor operation of the boiler with once-through utilization of the sorbent, without regeneration. However, since the increased waste disposal requirements resulting from once-through operation might be a problem in some cases, he felt that regeneration should continue to be investigated experimentally. Dr. Archer thus concluded that the first fluidized-bed boiler prototype could be built without provisions for regeneration.

Mr. Walker (Babcock and Wilcox), referring to the shortages of gas and oil envisioned in Professor Squires' keynote address, suggested that perhaps additional reserves will be discovered; historically, projections of gas and oil reserves have forecast shortages, but new finds have always been made which extend the projected reserve depletion date. Also, secondary recovery of oil from abandoned fields would be a potential source of oil, albeit at costs somewhat greater than current costs. Professor Squires (CCNY) agreed that perhaps up to 100 billion barrels of oil could be obtained by secondary recovery, and that much work might thus be justified in this area. However, he stated that, even if the oil reserves could be increased by a factor of 2, or even by a factor of 10, this oil would last no more than an additional 20 years at the current exponential increase in demand. Professor Squires also agreed that, in the past, new reserves of gas and oil have been located in time to prevent shortages, but he pointed out that shortages now appear to be actually occurring in the gas industry. Similar shortages might occur in the petroleum industry later. An increase in gas prices might result in increased exploration and gas field development, but Professor Squires doubted that the overall picture for natural gas could be significantly changed.

Mr. Locke (NRDC) felt that there will indeed be an increase in gas prices, especially as the demand for oil of small developing nations increases, and so puts a greater pressure on the U. S. oil and gas supplies.

Dr. Ulmer (Combustion Engineering) stated that the crisis is not the result of inadequate total energy supplies, but rather is a result of pollution control regulations. The

SO_2 emission limits have increased the demand for low-sulfur fuels, the supply of which is inadequate. The urgency of pollution regulations has forced an immediate allocation of resources toward the development of stack gas cleaning, with the result that these resources cannot be invested in longer-term solutions, such as fluidized-bed combustion.

Professor Squires concurred that the dates established by law for meeting emission standards are creating a sense of urgency which is forcing rapid, less-than-optimum solutions instead of permitting development of possibly more promising but longer-term technology. The standards for automobile air pollutant emissions are perhaps the strongest example of this problem.

Dr. Fisher (Chemico) commented that possibly industry needed the emission regulations to provide the incentive required to get control programs underway.

Dr. Fisher suggested that possibly Government regulations regarding the efficiency of fuel-burning plants should be considered.

Professor Squires feared that an approach involving specification of the end use of fuels will ultimately be imposed. He commented that a tax on pollutant emissions would be a suitable means for encouraging industry to control itself only if the time schedule for imposing the tax were realistic.

Mr. Walker asked Professor Squires how much time and money might be necessary to develop the overall "Coalplex" approach as rapidly as possible. Professor Squires could not estimate the required financial investment; he felt that it would be at least ten years before the approach could make a significant impact. He believed that, even if such a solution could be developed in ten years, it might be too late to prevent the forecasted energy shortages.

Dr. Archer referred to the tax which some utilities have placed on themselves in order to raise funds for sponsoring, through the Electric Power Research Institute, a research and development program which will be designed to advance the state of the art of power generation. This selfimposed tax was apparently instituted only after some members of the United States Senate had threatened to impose a political tax to provide funds for such an R&D program. Thus, Dr. Archer concluded that this tax might be an example indicating that industry may need at least a threat of Government involvement as an incentive to undertake this type of program.

Mr. Locke stated that, if the Government involvement has the effect of forcing a short-term solution, then interest in a longer-term solution may well be inhibited. For example, if utilities are forced to reduce pollutant emissions using stack gas cleaning techniques now, then there may not be as much incentive for developing longer-term techniques to reduce emissions in the future. Short term legislation should bear in mind the wider technological and commercial indications, and planning needs to consider the longer term too.

Professor Squires commented that the scientific community should do a better job of informing Congress regarding the status and potential of technology. A better-advised Congress would be better able to enact realistic legislation in this area.

Dr. Fisher suggested that perhaps the high costs of stack gas cleaning might give Congress an indication that alternate, longer-term processes should be considered. Professor Squires agreed, citing low-Btu gasification as an example of one concept that has received increased attention.

Dr. Reh (Lurgi) stated that the copper industry offers another example of a situation in which the urgency of pollution control regulations is compelling plants to rapidly install existing control technology instead of working towards improved future technology.

Professor Elliott (University of Aston) suggested that low-Btu gas will not be widely utilized in large central stations. Rather, he believed that the low-Btu "power gas" will be employed in small plants for central heating in the cities, while large stations will be nuclear.

Dr. Gorin (Consolidation Coal Co.) believed that, due to the operating nature of some envisioned low-Btu coal gasification processes, it would not be practical to employ these processes on the swing-load basis suggested by central power station application. Professor Elliott stated that the gasification plant could be operated as base load, even in a central power station application, provided that a heat storage concept such as that proposed by Stal-Laval were employed. Professor Squires added that he has been considering a pyrolysis scheme in which, by storing some of the fuel product, the pyrolizer would be continuously operated at full load, although only one-third of the output would be needed as base load with the other two thirds as swing load.

Dr. Gorin stated that the coal gasification process could be operated at full load, with peaking being accomplished utilizing liquid fuels obtained from the coal. Professor Squires said that that is the arrangement that he is considering.

Mr. Hangebrauck asked for comments regarding the fluidized-bed combustion concept in which the pressurized fluidized combustor is operated without steam-generating tubes in the bed. In such a system, the bed temperature would be controlled by utilizing a high excess air rate, and the off-gases would pass through a heat recovery boiler after having been expanded through a gas turbine. The CPU-400 incinerator program being conducted by Combustion Power Company utilizes such a system to burn municipal refuse.

Dr. Archer stated that such a system would require a larger bed, larger particulate removal equipment, and larger heat recovery boilers than would a system including tube surface in the combustor, due to the higher gas flows in the former system. Also, due to the relatively low temperature of the expanded gases leaving the turbine, the heat recovery boiler would have to be fired with supplementary fuel if it were desired to generate high-quality steam. Dr. Archer felt that steam of reasonably high-quality would be needed for high overall combined cycle efficiency. The refuse-burning CPU-400 system does not depend as heavily on high cycle efficiency as would a coal-burning power generation combined cycle system based on this "adiabatic" combustor principle.

Mr. Furlong (Combustion Power Co.) felt that coal combustion in the "adiabatic combustor" system is nearer term than is the pressurized combustion system in which the combustor does contain transfer surface, or than is a gasification system. Mr. Chapman (EPA) stated that the equipment needed to study the "adiabatic combustor"/combined cycle system is essentially available at the present time, in the form of the EPA-sponsored CPU-400 pilot plant, so that answers to questions regarding this system could be obtained on an early basis.

Mr. Walker commented that a system to generate power by burning waste material would conserve the reserves of fossil fuels. Mr. Harboe (Stal-Laval) suggested that such refuse-burning plants should be distributed in residential areas, as part of a district heating system; the energy from the plants could be stored during periods of low demand, and used for space heating when needed. Professor Elliott felt that such incineration/central heating systems could be installed on a household-by-household basis.

Mr. Hangebrauck asked the panel and other attendees what future they foresaw for fluidized-bed combustion processes.

Professor Elliott believed that such processes have a big future and that large fluidized-bed combustion plants with topping cycles would be built by the year 2000. Mr. Skopp (Esso Research & Engineering Co.)

asked Professor Elliott if he felt that a bottoming cycle might offer promise. Professor Elliott responded that a bottoming cycle would not increase plant efficiency, but could only reduce it.

Mr. Sullivan (Gilbert Associates) asked what plant efficiencies Professor Elliott foresaw in future years. Professor Elliott expressed the opinion that an overall efficiency of 55 percent could be achieved now, and that 60 percent could be reached in the future.

Mr. Hangebrauck asked for discussion of the potential of installing a coal gasifier to provide fuel for an existing conventional power plant.

Dr. Archer believed that the ultimate application of low-Btu gasification systems would be as part of an advanced power cycle, and that the use of low-Btu gas in conventional boilers would not be economical. Mr. Matthews (IGT) agreed. Mr. Locke pointed out that in the sequence from pressurized fluidized combustion via conventional atmospheric pressure combustion to gasification followed by combustion, the capital cost component of power send-out increased. It increased further with every additional process unit and heat exchanger added into the flow sheet; so also did operational inflexibility and maintenance costs. He pleaded that design ingenuity should be aimed to achieve simplicity rather than complexity in system scheming.

Mr. Walker stated that the use of a Lurgi gasifier has been considered for providing gas to an existing 750-MW plant. This analysis had indicated that the existing furnaces did not have sufficient wall surface to accommodate the larger burners that would be needed for the low-Btu gas. Thus Mr. Walker concluded that a boiler could be fired with such low-Btu gas only if the original unit design provided for such a fuel; an existing boiler could not be retrofitted. He agreed that perhaps a retrofit might be possible if the gas had an intermediate heating value, on the order of 500 Btu/scf.

Professor Elliott said that possibly an improved burner design would overcome the problems that Mr. Walker foresaw with low-Btu gas.

Mr. Hangebrauck asked if a boiler designed to burn coal would have to be derated if it were converted to low-Btu gas—asuming that improved burner design would allow low-Btu gas to be fired. Professor Elliott said that the boiler would have to be derated because the emissivity of the gas flame would be lower. He suggested that, if the gas burner were designed to give carbon formation, the emissivity could be increased and the amount of derating reduced.

Mr. Harboe commented that the envisioned advanced power cycle systems will be large, refinery-like complexes which not only will attract increased attention from local pollution control authorities, but which—if located near residential load centers—will have to be acceptable in appearance. He suggested that the utility industry, which would have to build and operate these plants, might be asked what systems they believe should be developed. Dr. Archer said that it might not be fair to ask the utilities to make decisions regarding future systems; they are already in the difficult situation of having to think years in advance when ordering a conventional plant.

Minutes submitted by:

D. B. Henschel

APPENDIX
ATTENDANCE LIST

(NOTE: To facilitate their identification, attendees are listed alphabetically together with the name of the organization they represent. The complete address of each organization represented at the Conference appears at the end of the list of attendees.)

LIST OF ATTENDEES

Name	Representing
Archer, Dr. David H.	Westinghouse
Bernard, Mr. A.	Babcock-Atlantique
Bryers, Mr. R.W.	Foster-Wheeler
Carls, Mr. E.L.	Argonne National Laboratory
Casapis, Mr. Michael	United Engineers and Constructors
Chapman, Mr. R.A.	EPA (Cincinnati)
Cole, Mr. W.E.	Pennsylvania State University
Copeland, Mr. G.G.	Copeland Systems
Corder, Mr. William	Battelle
Curran, G.P.	Consolidation Coal Company
Dickey, Mr. B.R.	Allied Chemical
Diehl, Mr. E.K.	Bituminous Coal Research
Dravid, Mr. A.N.	Shell Development
Ehrlich, Mr. Shelton	Pope, Evans and Robbins
Elliott, Professor Douglas E.	University of Aston (England)
Fisher, Dr. John V.	Chemical Construction
Furlong, Mr. D.L.	Combustion Power
Giberti, Dr. R.A.	Peabody Coal
Glenn, Mr. Roland D.	Combustion Processes
Godel, Mr. Albert A.	Societe Anonyme Activit
Gorin, Dr. Everett	Consolidation Coal
Hangebrauck, Mr. R.P.	EPA (Durham)
Hanway, Mr. John E., Jr.	Chicago Bridge and Iron
Harboe, Mr. Henrik	Stal Laval (England)
Henschel, Mr. D.B.	EPA (Durham)
Highley, Mr. John	National Coal Board (England)
Hoke, Dr. R.C.	Esso Research and Engineering (Linden)
Holmes, Mr. John M.	Oak Ridge National Laboratory
Jonke, Mr. A.A.	Argonne National Laboratory
Keairns, Dr. Dale L.	Westinghouse
Lewis, Mr. P.S.	Bureau of Mines (Morgantown)
Locke, Mr. H. Brian	National Research Development

List of Attendees (Cont)

Name	Representing
Matthews, Mr. C.W.	Institute of Gas Technology
Moss, Dr. Gerty	Esso Petroleum (England)
Newby, Dr. R.A.	Westinghouse
Nutkis, Mr. M.N.	Esso Research and Engineering (Linden)
Olson, Mr. G.A.	Electric Research Council
O'Neill, Mr. E.P.	Westinghouse
Pell, Dr. Melvyn	Consolidation Coal
Rakes, Mr. S.L.	EPA (Research Triangle Park)
Reh, Dr. Lothar	Lurgi
Reintjes, Mr. Harold	Petrocarb
Rice, Mr. R.L.	Bureau of Mines (Morgantown)
Robson, Dr. F.L.	United Aircraft
Schmidt, Dr. H.W.	Lurgi
Sears, Dr. John T.	West Virginia University
Shackelford, Mr. J.M.	EPA (Arlington)
Skopp, Mr. Alvin	Esso Research and Engineering (Linden)
Spector, Mr. Marshall L.	Air Products and Chemicals
Squires, Dr. Athur M.	CCNY
Stewart, Mr. J.T.	Bituminous Coal Research
Stewart, Dr. O.W.	University of Kentucky
Sullivan, Mr. Leo	Gilbert Associates
Sverdrup, Mr. E.F.	Westinghouse
Turner, Mr. P.P.	EPA (Research Triangle Park)
Ulmer, Dr. Richard C.	Combustion Engeneering
Vance, Mr. S.W.	Dorr-Oliver
Vogel, Mr. G.J.	Argonne National Laboratory
Walker, Mr. James B., Jr.	Babcock and Wilcox
Wright, Dr. S.J.	National Coal Board (England)
Zimmerman, Mr. R.D.	Gulf General Atomic
Zitzow, Mr. Uwe	TVA

LIST OF ORGANIZATIONS REPRESENTED

Name (Represented by)	**Address**
Air Products and Chemicals, Inc. (Mr. Spector)	P.O. Box 538 Allentown, Pennsylvania 18105
Allied Chemical Corp. (Mr. Dickey)	P.O. Box 2538 Idaho Falls, Idaho 83401
Argonne National Laboratory (Mr. Carls, Mr. Jonke, Mr. Vogel)	9700 South Cass Avenue Argonne, Illinois 60439
Babcock-Atlantique (Mr. Bernard)	48 Rue La Boetie Paris, 8^e, France

List of Organizations Represented (Cont)

Name (Represented by)	Address
Babcock and Wilcox Co. (Mr. Walker)	20 South Buren Avenue Barberton, Ohio 44203
Battelle Columbus Laboratories (Mr. Corder)	505 King Avenue Columbus, Ohio 43201
Bituminous Coal Research, Inc. (Mr. Diehl, Mr. J. Stewart)	350 Hochberg Road Monroeville, Pennsylvania 15146
Bureau of Mines (Mr. Rice, Mr. Lewis)	P.O. Box 880 Collins Ferry Road Morgantown, West Virginia 26505
Chemical Construction Corp. (Mr. Fisher)	320 Park Avenue New York, New York 10022
Chicago Bridge and Iron Co. (Mr. Hanway)	Route 59 Plainfield, Illinois 60544
City College of the City University of New York (CCNY) (Dr. Squires)	245 West 104th Street New York, New York 10025
Combustion Engineering, Inc. (Dr. Ulmer)	1000 Prospect Hill Road Windsor, Connecticut 06095
Combustion Power Co. (Mr. Furlong)	1346 Willow Road Menlo Park, California 94025
Combustion Processes, Inc. (Mr. Glann)	515 Wythe Street Alexandria, Virginia 22314
Consolidation Coal Co., Inc. (Mr. Curran, Dr. Gorin, Dr. Pell)	Library, Pennsylvania 15129
Copeland Systems, Inc. (Mr. Copeland)	120 Oakbrook Mall, Suite 220 Oakbrook, Illinois 60521
Dorr-Oliver, Inc. (Mr. Vance)	77 Havermeyer Lane Stamford, Connecticut 06904
Electric Research Council (Mr. Olson)	90 Park Avenue New York, New York 10016
Environmental Protection Agency (Mr. Shackelford) (Mr. Hangebrauck, Mr. Henschel, Mr. Rakes, Mr. Turner)	Office of Research and Monitoring Xerox Building 1901 N. Fort Myers Drive Arlington, Virginia 20460 National Environmental Research Center Research Triangle Park, N.C. 27711

List of Organizations Represented (Cont)

Name (Represented by)	**Address**
Esso Petroleum Co., Ltd. (Dr. Moss)	Esso Research Centre Abingdon, Berkshire, England
Esso Research and Engineering Co. (Dr. Hoke, Mr. Nutkis, Mr. Skopp)	Government Research Laboratory P.O. Box 8 Linden, New Jersey 07036
Foster-Wheeler Corp. (Mr. Bryers)	12 Peach Tree Hill Road Livingston, New Jersey 07039
Gilbert Associates, Inc. (Mr. Sullivan)	525 Lancaster Avenue Reading, Pennsylvania 19603
Gulf General Atomic, Inc. (Mr. Zimmerman)	P.O. Box 608 San Diego, California 92112
Institute of Gas Technology (Mr. C.W. Matthews)	3424 South State Street Chicago, Illinois 60616
Lurgi Gesellschaft Fuer Chemie und Huttenwesen mbH (Dr. Reh, Dr. Schmidt)	Lurgihaus, Gervinusstrasse 17/19 Post Fach 9181 6000 Frankfurt (Main) Germany
National Coal Board (Mr. Highley, Dr. Wright)	Hobart House, Grosvenor Place London S.W. 1, England
National Research Development Corp. (Mr. Locke)	P.O. Box 236 London S.W., 1E 6SL, England
Oak Ridge National Laboratory (Mr. Holmes)	Process Design Section P.O. Box X Oak Ridge, Tennessee 38730
Office of Coal Research	U.S. Department of Interior Washington, D.C. 20240
Peabody Coal Co. (Dr. Giberti)	301 North Memorial Drive St. Louis, Missouri 63102
Pennsylvania State University (Mr. W.E. Cole)	Combustion Laboratory University Park, Pennsylvania
Petrocarb, Inc. (Mr. Reintjes)	250 Broadway New York, New York 10007
Pope, Evans and Robbins (Mr. Ehrlich)	515 Wythe Street Alexandria, Virginia 22314
Shell Development Corp. (Mr. Dravid)	P.O. Box 481 Houston, Texas 77001

List of Organizations Represented (Cont)

Name (Represented by)	**Address**
Societe Anonyme Activit (Mr. Godel)	66 Rue D'Auteuil Paris, XVI^{e}, France
Solid Waste Research Lab. (Mr. Chapman)	5555 Ridge Avenue Cincinnati, Ohio 45268
Stal Laval, Ltd. (Mr. Harboe)	Villiers House 4 Strand London, Wc2N 50H, England
Tennessee Valley Authority (Mr. Zitzow)	720 Chattanooga Bank Building Chattanooga, Tennessee 37401
University of Aston at Birmingham (Prof. Elliott)	Gosta Green Birmingham, B4 7ET, England
University of Kentucky (Dr. O. Stewart)	Lexington, Kentucky 40506
United Aircraft Research Lab. (Dr. Robson)	East Hartford, Connecticut 06108
United Engineers and Constructors, Inc. (Mr. Casapis)	1401 Arch Street Philadelphia, Pennsylvania 19102
West Virginia University (Dr. Sears)	Chemical Engineering Dept. Morgantown, West Virginia 26506
Westinghouse Electric Corp. (Dr. Archer, Dr. Keairns, Dr. Newby, Mr. O'Neill, Mr. Sverdrup)	Research and Development Center Beulah Road, Churchill Borough Pittsburgh, Pennsylvania 15235

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